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BIOMEDICAL AND BEHAVIORAL SCIENCES

No. 2

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CONTENTS PAGE

CONTENTS (Continued) Page

ENVIRONMENTAL HAZARDS

RESULTS AND PROBLEMS OF THE MEDICAL SURVEILLANCE OF PERSONS WORKING WITH IONIZING-RADIATION SOURCES IN THE USSR

New York City PROCEEDINGS OF THE FOURTH INTERNATIONAL CONFERENCE ON THE PEACEFUL USES OF ATOMIC ENERGY in Russian Vol 11, 1972 pp 225-241

/Article by A.A. Letavet, A.K. Gus'kova, P.P. Lyarskiy and P.I.Moiseytsev USSR Academy of Medical Sciences, Moscow. E.I. Vorob'yev, USSR Ministry of Health, Moscow/

/Text/ Results of 15 years of observations indicate that radiation doses received by personnel of scientific and medical institutions, operators of reactors and of particle accelerators and of radiographs under ordinary conditions presently constitute one-half to one-tenth the maximum permissible dose. The intake of significant quantities of radioactive substances is practically impossible. Materials involving 6000 persons were subjected to detailed dynamic clinical-physiological study in comparison with adequate control groups.

The most adequate (for the given level of dose parameters) observations which permit a quantitative evaluation of the state of the resulting functions of basic critical organs and systems (hemodynamics and its regulation, cardiological and cytological methods and others) are evaluated. Situations in which there is a possibility of increased, most frequently variable radiation from external sources (transportation and reloading of sources, gamma-flow detection under field conditions, investigative installation of the critical mass of reactors) and the clinical manifestations characteristic of them are analyzed.

There are formulated fundamentally new questions of present-day medical examinations, which permit: clarification of the phenomenology of the initial adaptive syndrome of the reaction to radiation, acting in a complex with other occupational factors; the evaluation of the possibility and mechanisms of reparation after previously existing more intense radiation,

especially at dose levels, which cause sub-clinical lesions; substantiation of adopted standards not according to direct reactions but according to the criterion of remote after-effects of radiation.

Fundamental bases of legislation adopted in the USSR for protection of persons working with radiation sources in the national economy in comparison with the analogous International Convention No.115 MOT /International Labor Organization/ are listed and analyzed. Means of improving systems of medical and hygienic observations and possibilities of purposeful selection and summarization of information collected in different countries according to a unified program and necessary for ensuring extensive safe use of radiation for peaceful purposes are evaluated.

The steady increase of the number of specialists working with sources of ionizing radiations and also the extension of the terms of observation provide some results of medical-hygienic studies in this area and suggest means for further study of this question.

Nearly 15 years ago A.A. Letavet /1/, at the First Conference on the Use of Atomic Energy for Peaceful Purposes, in Geneva in 1955, described a system of governmental legislation and practical recommendations adopted in the USSR in this period in respect to types of work which involve radiation hazards. Later, the Soviet Union ratified Convention No. 115 of ILO concerning the protection of workers from ionizing radiation and confirmed the closeness of the principal positions of our country with all basic positions of this convention.

Thus, there now exists a real possibility for evaluating the basic practical results of use of actual scientific information obtained in the process of medical observation of workers exposed to radiation sources.

In this report, naturally, we are relying predominantly on experience accumulated by scientific research and practical medical institutions of the Soviet Union and constantly comparing it with analogous materials obtained by scientists of other countries.

A distinguishing feature of this period of production and use of radiation sources in different sectors of industry, medicine and scientific research in all countries is the steady increase in the number of persons who come into contact with radiation sources and the kinds of contacts with radiation, which are 150-200 percent respectively of the levels existing 10-15 years ago in the highly developed countries. The overall volume of use of radioisotope devices in the USSR has increased more than 3-fold in the last 5 years alone.

Analysis of reports of the USSR sanitation and epidemiological service and also similar departments of Canada (1963, 1966) /2,3/, France (1967) /4/, USA (1965) /5/, analysis of reports of the TsERN [CERN] (1969) /6/ and, finally, the most complete summary presented by the Scientific Committee of the United Nations in 1970 /7/ on the problem of the percentage of the population coming into contact with radiation in their work, reveals the following important facts.

The percentage of persons occupationally exposed to radiation constitutes, at present, 1-2 percent of the total population, depending upon the level of development of the country. At the same time, the number of persons working in the atomic industry is 4-8 times less than those who use sources of radiation. The medical and science sectors, with the vast diversity of kinds of contact due to the nature of their activity, lead all other sectors in this regard. Many persons work with reactors intended for different purposes and their number is increasing. The general trend of increase of numbers and the relationship between individual occupational groups in different countries is closely related, which gives special importance to the following conclusions and recommendations /6, 10/.

Another important feature of the present period is the significant universal reduction of level of radiation achieved in the USSR and in other countries. In 90-99 percent of the workers, this quantity is 1/3 less than the accepted maximum permissible value of occupational effect. Only in 4-9 percent of the workers does occupational radiation constitute 1.5 - 5 R per annum from external sources and it exceeds this level in less than 1 percent.

Mean values, according to data of the cited report of the Scientific Committee of the UN /7/ for the majority of the sectors vary from 0.07 to 0.59 R/annum.

Analysis of numerous data accumulated by hygienists of the Soviet Union /8-10/ show that, for basic occupational groups of research reactors personnel, of charged particle accelerators attendants working on radiation monitoring and regulating devices, the level of radiation is, as a rule, 3-5 times lower than the accepted maximum permissible dose (PDD). Only in persons in certain occupations, involved in repair operations, who perform gamma-flaw detection and well logging under field conditions and also those working on experimental nuclear filters, does the dose of radiation approximate the permissible maximum, constituting from 2 - 4.5 roentgen equivalent man/year. (REM).

Many years of hygienic observations of work conditions during use of different types of nuclear reactors in the USSR indicate that they are

presently completely satisfactory. Under stable regimes of operation, the power of gamma-radiation constitutes 0.01 - 0.02 microR/second; the flows of thermal, intermediate and fast neutrons are 20-70; 10-15; 5-10 neutrons/cm^2·seconds, respectively. The content of active short-lived aerosols in the air of the basic production areas does not exceed 10^{-14} 10^{-13} Ki/liter (oxygen utilization factor) and that of long-lived - 10^{-15} Ki per liter.

The overall dose of radiation of personnel who service reactors involved in scientific research, according to the sum of all components of radiation (gamma, beta and neutrino), does not exceed 0.5 - 1 rems/year and only for a very small number of persons in the central hall, occupied in repair of technological channels and in working out the reactor regime, reaches 2.5 - 4.5 rem /year.

Analogous values (0.5 - 4.0 rems/year) are obtained during ongoing operation of different types of charged particle accelerators (synchrophasotron, synchrocyclotron, betatron), of powerful isotope gamma-devices, used widely for many radiochemical processes and "radiation" sterilization and also of some other highly-active sources of radiation (different types of research filter circuits used widely for physical and physical-chemical studies and analysis of the structure of substances).

The most unfavorable work, at present, from the hygienic point of view, is work involving industrial flaw detection, especially that which involves the use of portable apparatus. Exposure of personnel from these devices during some forms of manipulation is presently at the limit of permissible levels (3-5.5 rems/year). In stationary gamma-flaw detection, the exposure level is, at present, reduced to 0.8 - 1.5 rems/year.

Labor conditions of employees of medical roentgeno-radiological institutes are extremely diverse today. Figure 1 shows the change of the peripheral blood in roentgenologists, related to the formation of overall doses of radiation. Analysis of the dose levels for different types of procedures and periods of time, for which are possible accumulation of different overall values, in comparison with the potential danger of the rise of unfavorable remote after-effects, was undertaken by V.Ya. Golikov /11/. It was shown that, for the overwhelming majority of workers, with the exception of persons conducting special roentgenological and radiotherapeutic procedures (trochoscopy, angiopulmono-cardiography, stowing applicators and caring for radio-oncological patients), the overall dose and locally absorbed doses are lower than those whose effect could possibly lead to development of local changes of the integument (800-1200 rads) or to such remote unfavorable after-effects as an increase in frequency in the cases of leukemias and a shortening of life.

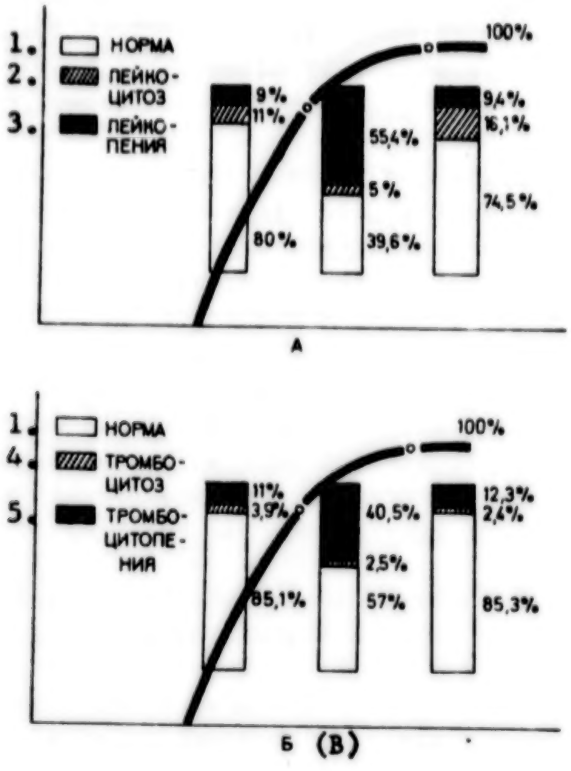

Figure 1. Assessment of radiologists according to indicators of the peripheral blood in different periods of work in comparison with the dynamics of formation of the overall dose exposure. On the curve is the relative dose value (in percent of the overall dose). In the columns is the relative number of persons with reduced, normal and increased number of leukocytes (A) and thrombocytes (B) in the peripheral blood.

Key:
1. Normal
2. Leukocytosis
3. Leukopenia
4. Thrombocytosis
5. Thrombocytopenia

In connection with the above mentioned general reduction of the radiation levels and the expansion of the area of use, there is being noted, in all countries, a gradual growth of the relative significance of non-radiation factors in the overall complex of effects of work conditions on the state of health of persons working with radiation sources. This should be considered both in the medico-hygienic examination and also during evaluation of the observations, especially the remote effects.

We must also note the appreciable increase in the average age of persons professionally associated with radiation inasmuch as changes in the state of their health, caused by physiological aging, intercurrent diseases and also some individual physiological and pathological states inherent to persons of different ages and sex, becomes more meaningful.

In standard documents adopted in past years in individual countries, the maximum permissible level of radiation was lowered gradually. These recommendations became more stable after 1960 and were not reconsidered to any great extent /12-14/.

Real possibilities of significantly reducing occupational radiation loading arose in the process of mastery of the technology. Thanks to this, even persons remaining at their previous work posts were subjected to noticeably reduced radiation levels and the overall dose is very unevenly distributed in the course of their long occupational activity /15-17/.

Differentiated analysis conducted by E.A. Denisova /18/, (1970) on individual groups of workers established that this excess never involves persons working on research and power reactors and the overall values of doses among them for the entire work time does not exceed 30 R. Among two-thirds of accelerator workers with a long period of service, the combined doses, at present, reach 80-100 R but, for more than one-half of the roentgenologists, they exceed 100 R.

Among medical radiologists who began work in the USSR in the last 10 years /19-23/, the combined doses, as a rule, are lower than 15-30 R with relatively uniform distribution for the entire period of their professional activity.

Reconstruction of distribution of dose loadings in time for all of these persons and especially for persons being exposed to combined radiation effect from different sources (contact with radium compounds, thorium and others) seems to us to be much more important than formal analysis of effects conducted only according to comparison with combined doses of occupational radiation.

In respect to the combined effect of some factors, a methodology of research involving a significant revision of some standing ideas is being formulated. It is enough to point out the complex evolution of opinions concerning the nature of diseases developing among miners who come into contact with ores containing admixtures of radioactive substances or in persons who produce and apply permanent luminous compounds containing radium-226, mesothorium and other substances to articles /24-27/. The basis of future extremely interesting studies in this direction should be improvement of methods of quantitative characteristics of each factor in different periods of observation, the creation of adequately precise methods of judgment concerning the function of basic damaged organs and systems, precise representation of the biological effect of individual components, constant clinical and radiometric comparisons with dose levels established for the given organ by an entire complex of factors and the establishment of the comparative significance of these or those components in different periods of observation.

We must point out that the concept "critical organ" in its proper interpretation remains an important base of reference in this kind of investigation.

It is interesting that this concept, being generated within the framework of the study of the biological effect of radioisotopes with selective distribution /28-29/, is beginning to be used effectively also for evaluation of changes, caused by radiation from external sources, especially with the unequal distribution of energy inside the body /30-33/.

Everything indicated above determines the following practical important considerations. The possibility of detection of developed clinical syndromes of chronic radiation sickness among occupational contingents presently is becoming highly improbable (A.K. Gus'kova, 1970).

Much rarer are cases of severe, general, acute radiation injuries which were discussed in reports of appropriate departments in the USA /5, 34/, Canada /35/ and England. Summary data were also presented at a symposium on accidents in Vienna in 1969 /36, 37/, in works by L.L. Sokolina /38/ (1970) and also in some other publications /39/.

A few cases of recognition of chronic radiation sickness involve limited numbers of persons who began to work many years ago. As a rule, there was found, in them, mild and obliterated forms of disease in the period of complete or partial recovery /40-43/.

Individual acute injuries recorded in publications in recent years show a predominance of cases caused by irregular radiation /44, 45/ or local forms;

in 9/10 of the cases there is evidence of serious violation by the personnel of the rules of work and not the consequence of defects of technology of the equipment, /46-53/.

The predominance of local changes is characteristic on the whole also for the contingent of patients of radiation sickness primarily registered in the last 10 years. Sub-clinical forms of these injuries are most frequent in materials of dispensary observation for certain occupational groups /20, 52, 54, 55/.

The clearly marked tendency in recent years to the predominant effect from external sources of radiation and, in any case, the more frequent determining role of the latter in comparison with the entrance of radioactive substances inside in the biological effects being observed is very interesting /38/.

If the reduction of frequency of cases of chronic and acute radiation sickness and reduction among them of the percentage of severe injuries is completely uniform for all countries, then this, regrettably, is not the case in respect to so-called accidents or emergency occupational situations as a whole. The frequency of cases of unforeseen radiation of personnel in larger than normal doses without leading to the rise of defined forms of radiation injuries but which require qualified expertise and rational labor arrangement is being maintained at the previous level or even increasing somewhat (summary data, presented at the Symposium on Accidents in Vienna in 1969, materials concerning incidents with radium in the USA /56/ and others).

The distribution of frequency of cases of exposure to high doses of radiation (without clinical manifestations of reactions, with the presence only of local and, of course, overall radiation injuries) is extremely characteristic and similar in materials presented by different countries. Among 76 cases of high radiation doses, summarized by L.L. Sokolin (1969) /38/, only one of the sufferers experienced acute radiation sickness of the 1st degree from non-uniform radiation. There were observed 23 cases of local injuries and, in the others, there occurred only an increase of the maximum levels of radiation without obvious clinical manifestations.

It is important to note that in some occupation related situations, (loss of the radiation source) a possible effect of emissions involves equally individuals from the population of different ages who do not have any idea about the degree of radiation hazard /34-38/. The tragic effects of such situations in one Mexican family has been reported many times in the periodical press (Gonzales and Berumen, 1963, /57/).

Results of analysis of all published clinical observations permit us to say that during an overall exposure dose of general radiation of 70-100 R and an intensity of radiation of less than 0.05 R/day, it is not possible to observe obvious symptoms of injury, even on the part of the most sensitive organs and systems (hematogenesis, crystalline lens and others). This refers to cases of momentary emission in the indicated dose, keeping in mind the evaluation of the dynamics of shifts in each individual subject.

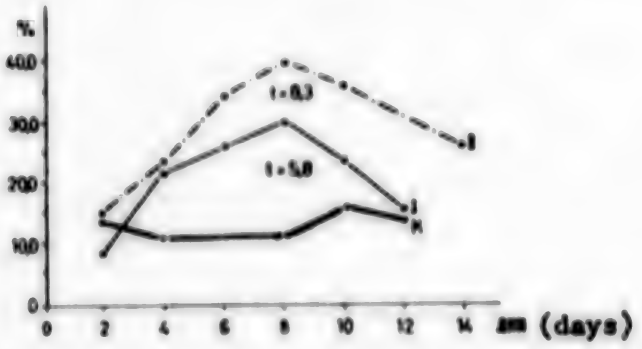

Figure 2. Frequency of the syndrome of neurocirculatory dystonia in different periods of observation in the control group (K) and in persons, who are subjected to occupational radiation in overall doses of 20-50 REM (I) and 70-100 REM (II).

However, even beginning from combined doses of 20-50 R (and more regularly during achievement of values of 70-100 R during use of special methods of clinical-physiological study) there can be noted in larger groups, in $\sim$ 4.5 of the cases, development of a clinical syndrome of expanded physiological reactivity of systems, which ensure adaptation of the organism to the conditions of the external environment. In its development is discerned a combination of effect of some factors which facilitate impairment of adaptational mechanisms of neuro-vascular regulation; neuropsychic or somatogenic asthenia in those above 40 years of age. Later, even during continuing exposure in doses which do not exceed 1-3 R/year, there occurs, in most cases, a smoothing out of manifestations of the indicated reactions, evaluated by us as orientational-adaptational reaction to the complex of all factors of the environment, including radiation. Merely as an example, there occurs a strengthening or even an inincrease of regulatory shifts in 17-24 percent of those examined. A hypotonic neutrocirculatory dystonia syndrome is being formed. In the other 3/4 to 4/5, there occurs a gradual convergence of frequency of shifts of individual indicators and of the syndrome as a whole with the limits of their variability in adequate control with only insignificant change during dynamic observation in the course of 6-8 years, being encountered in 12-14 percent of the persons examined.

This position may be shown by an appropriate illustration of the frequency of individual shifts in persons, who are subjected to radiation and in adequate control, during dynamic observation of them (Figure 2).

The facts presented above indicate how much the situation concerning persons occupationally associated with radiation was changed. Other and basic trends of scientific-practical and research activity must be initiated. The detection and diagnosis of developed forms of radiation sickness and the realization of therapeutic-prophylactic measures in respect to direct reactions to radiation can no longer be considered to be the basic task of periodic medical examinations as it was in the period of establishment of the atomic industry and in the initial period of use of radiation in the national economy /1/.

At the same time, definite progress in knowledge is being achieved and considerable practical experience is being accumulated in the area of the clinic of radiation reactions in man which permit more skillful and sound resolution of many disputed problems. Basic positions of this study, in particular, are based on materials of special clinical-physiological observations conducted by the Institute of Labor Hygiene and Occupational Diseases of the Academy of Medical Sciences and some other scientific collectives of more than 6000 persons working with radiation in medicine, at research reactors, particle accelerators, in industrial gamma-roentgenography and those having contact with radium compounds and other radioactive substances, being used in industry, agriculture and scientific institutions.

The basic results of this section of medical-hygienic study are:

a) detailed characteristics and evaluation of the significance of basic radiation factors in clinical manifestations in persons in occupations involving contact with radiation sources;

b) determination, on the basis of numerous clinical-hygienic comparisons, of the principles of dispensary observation of these persons, the real possibilities of the rise and the clinical variants of reactions caused by occupational radiation effect;

c) the accumulation of information concerning clinical criteria on the basis of maximum permissible levels of occupational radiation for groups of workers (according to the criterion of direct effects) and the development of a methodology of further study of this problem;

d) creation of a general pathogenetic classification and characteristics of basic forms of radiation sickness in man in different periods of its course;

e) assessment of the variability of standard indicators thanks to expansion of contingents and increase of the courses of observation for the so-called control groups, completely necessary for producing reliable information concerning regularities of shifts detected in persons subjected to radiation and the establishment of their connection with the effect of radiation /58-61/.

It is especially important to emphasize, in this group, the developments of the publications of V.V. Sokolov and I.A. Gribova (1969) concerning the indicators of the peripheral blood and the bone marrow of healthy persons, which include personal observations and critical analysis of extensive data from the literature concerning norms of blood cells, their variability and precision of the methods themselves of counting them.

E.A. Denisova's observations (1970) concerning the dynamics in the course of 6-8 years of basic indicators of the blood circulation and its regulation, studied with the help of a wide spectrum (more than 15) of methods among 220 healthy persons, deserve attention and require further development. There are shown typical (for 78-83 percent of those examined), boundaries of variations of basic indicators and courses when there appear in them marked deviations caused by an increase in age.

Significant support in the evaluation of the conditions of the eyes and, especially, the crystalline lens of persons subjected to the effect of radiation in the USSR, came from basic research in the dynamics of shifts in healthy persons of different ages, including those subjected to the effect of radiation. These studies were conducted in the USSR by N.A. Vishnevskiy and his students V.I. Abdulayeva, V.N. Stiksova, D.N. Kotova, E.A. Ivanova and Ye.N. L'vovskaya /62, 63, 64/ during 1959-1970.

This is illustrated by the most indicative graphs from extensive materials of control studies of the eye, including dynamic observations for the extent of 7 years for 200 healthy persons in comparison with analogous information for persons subjected to the effect of radiation. In these studies, it was shown that, with a sufficiently large number of observations (100's and 1000's of persons examined), the control indicators for persons of the same age group, indicate practically complete identity in the mean values of the force of accommodation, the dimensions of the physiological field of vision, the degree of dilatation of the pupils upon introduction into the conjunctival space of a 1 percent solution of homotropin, the value of the intra-ocular pressure and the pressure in the central artery of the retina and also the frequency of deviations in the condition of structures of the anterior section of the vessels of the eye and the crystalline lens. An increase in age of 5 years and more leads to reliable narrowing of the caliber and to an increase of pressure in the retinal vessels. An increase in the frequency of spot opacification

in all layers of the crystalline lens is seen in persons of the control group as early as after 3-5 years of observation with the use of a slot lamp but is not ascertained in these same periods in respect to the frequency of changes under the posterior capsule (Figure 3).

A distinction of the group of persons subjected to occupational related radiation in doses near PDD /maximum permissible doses/ from gamma-sources was the reliable tendency toward a reduction of pressure in the central artery of the retina (TsAS) upon attainment of overall exposure radiation doses of the order of 70-100 R (Figure 4).

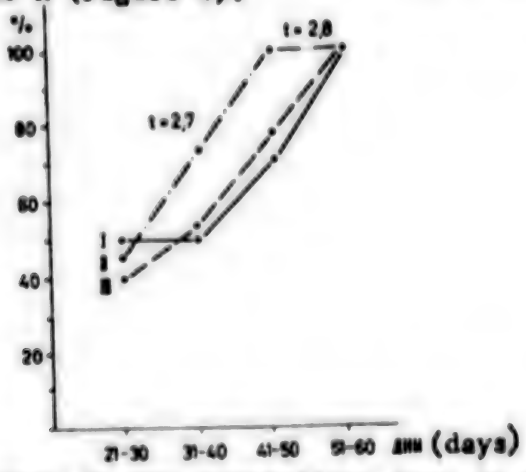

Figure 3. Frequency of changes in the crystalline lens in persons of different ages in the control group (I) in comparison with data concerning persons subjected to the effect of radiation in the range of 70-100 R (II) and 150-400 R (III).

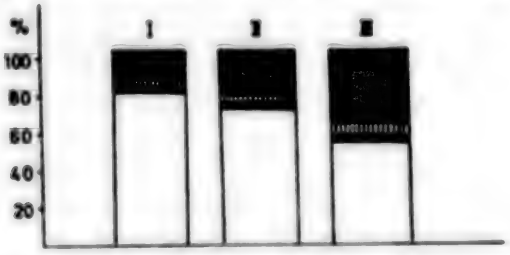

Figure 4. Frequency of cases of change of pressure in the artery of the retina in the control group (I) after radiation exposure in the overall dose of 70-100 R (II) and 150-400 R (III).

- Normal pressure in the central artery of the retina
- High pressure in the central artery of the retina
- Low pressure in the central artery of the retina

After radiation exposure in a dose of 150-400 R there was observed (more regularly in persons in the older age groups) some increase of frequency of spot opacities in vacuoles under the posterior capsule of the crystalline lens. There also appeared a certain tendency to acceleration of signs of physiological involution of the eye including a greater frequency of age-related cataracts than in the adequate control.

Only investigations of this nature can, in our opinion, provide an argued evaluation of the diagnostic significance of this or that shift and may be recommended representative courses of observation and proper evaluation of the contribution of occupation related radiation effect in the complex of effects acting on the dynamics of these or those deviations in the appropriate groups of persons.

Everything stated above indicates that a basic question at present is the creation of a reasoned system of purposeful medical examination which includes all persons who come into contact with radiation in their occupation.

It is advisable to differentiate the contingents and to determine the necessary volume, terms of observation and also possible yield of scientific information on the following bases.

For the significant part of workers, exposed all year to a radiation dose level of less than 1/3 the presently accepted PDD, there is, basically, no necessity (as is provided for by norms of radiation hazard) in a specialized examination to reveal indications of effects of radiation. On the first plane, for this category of persons, there emerge problems inherent in the principles of prophylactic observation adopted in the USSR and pursuing general medical purposes of prevention and timely treatment of some diseases, including those which prevent further work with radiation. The accumulation of comparative information on individual sectors of industry with a sufficiently large number of contingents and the study, among them, of the possible effect of low levels of radiations on general morbidity, mortality and the course of different general pathological and physiological processes (infectious diseases, tumors, pregnancy, age, the state of health of children, etc.) is of scientific interest.

Orientational counting of dose levels and forms of effect in this contingent (osteotrophic or hepatotrophic radiation, the overall external radiation, the effect on the skin of sources with low energies of generation) will permit further differential analysis of the contribution of the non-specific effect of radiation in the rise of syndromes of injury of critical organs and structures of man which syndromes are polyetiological in origin.

For the group of persons with radiation dose level from 1/3 to 1 PDD and near to PDD, it is advisable to maintain, in the light of present-day experience, more extensive differential observations once a year according to the radiation profile for the purposes of:

a) objective diagnosis of sub-clinical forms of reaction to the effect of radiation (mainly as a stimulus) and establishment of the possible effect of these shifts on the general and special capacity to work of personnel. There is also required the increase of precision and the possible connection of these shifts with processes being reflected under large overall doses of narrower effect of radiation as the damaging agent;

b) further clinical substantiation of accepted PDD of radiation dose according to the criterion of both direct and remote unfavorable effects of the occupational effect of radiation;

c) prevention of remote after-effects of radiation, the relatively low levels of which do not lead to noticeable direct effects.

For groups of persons, who were subjected, in the past, to radiation in doses which exceed the presently accepted permissible doses, who are suffering from radiation sickness or who are receiving a high dose of unforeseen radiation, very careful examination is performed at times and in amounts, dictated by clinical indications and also by the nature of the radiation (dynamics of formation of the dose load and distribution of absorbed energy inside the body).

Scientific information in this category of persons may provide important information:

a) on the dynamics of processes of radiation injury and recovery in different organs and systems of man;

b) on the nature and frequency of remote after-effects and outcomes after irradiation in a wide range of doses;

c) on the scientific argumentation of important practical problems of threshold levels of injurious doses;

d) on possibilities of rational labor layout and use of occupational classification of persons, who were previously subjected to a high dose of radiation or who suffer from different forms of radiation sickness.

In the USSR, there are some legal documents which regulate the basic aspects of ensuring radiation safety and some special recommendations are created concerning the organization of medical-hygienic observation in different sectors of the national economy. However, the improvement of legislation and its scientific substantiation are presently priority items in the work of leading scientific research institutions of the Soviet Union and its national commission on radiation protection. In addition to general acts of legislation concerning this or that aspect of human labor, there were adopted in recent years in the USSR some departmental rules and also particular methodical instructions for individual sectors, most complex situations or numerically important occupational groups.

This kind of material helps practicing physicians who conduct sanitation inspection and medical surveillance of persons working on reactors, accelerators, in medical and industrial gamma-graphy and radiography, with exposed isotopes of gold, phosphorus, iodine radium and others. They include, as a rule:

a) a brief written reference concerning the volume of information accumulated concerning a given occupational group;

b) typical characteristics of the level of influential factors and their possible biological action;

c) evaluation of potential sources of possible high radiation dose of a given professional group;

d) selection of adequate and accessible methodical means of study according to the real level of doses, typical radiation and also with norms of variability of recommended indicators in healthy persons;

e) description of clinical features which are possible in cases of high radiation from a given type of sources of forms of radiation sicknesses and reactions of individual organs and systems;

f) instructions on rational labor layout and expertise in cases of occupation related diseases and diseases not related to the occupation of a given contingent;

g) most advisable forms of registration, calculation and development of the data obtained.

As was indicated above, considering the relative rarity of such cases, the urgency of accumulating information collected into a unified plan and simultaneous rapid and operational, qualified decision making, the so-called "accidents" and 'emergency cases' in the area of producing and

using radiation sources require special clarification. Two instructional-methodical documents which include organizational formulation of a system of investigation and principles of rendering emergency assistance and prophylaxis measures in cases of high doses of radiation from internal and external sources were prepared by a group of scientific research institutes for these purposes in the USSR and were adopted and confirmed by the Ministry of Health.

The necessary scientifically reasoned refinement of definition concerning the practice of expertise and the preferential protection of persons working with sources of ionizing radiations in the USSR are also introduced into a general document concerning preliminary (upon beginning work) and periodical medical examination of workers, which document is confirmed by Decree of the USSR Ministry of Health No. 400 of 30 May 1960 with concurrence of the secretariat of the VTsSPS /All-Union Central Council of Trade Unions/. A list of occupational diseases, confirmed by the USSR Ministry of Health and VTsSPS on 25-26 February 1970 was determined. Instructions for its use include not only acute but also chronic radiation sickness and local radiation injury (p 19a, b, c). The possible connection between the effect of radiation and certain forms of tumors of the skin, bones and lungs (p 12, c, d and e) and also radiation cataracts (p 21a) is legally contended.

At the same time, there is urgent need for further improvement of legislation in the USSR in connection with the rapid growth of knowledge in the area of radiation medicine and the undeviating expansion of the sphere of use of radiation in the economies of all highly developed countries.

Most vital aspects in the plan of further scientific investigations on problems of radiation, intended to make a specific economic contribution to the use of radiation for peaceful purposes are:

a) Selection of more adequate quantitative methods of objective registration of sub-clinical shifts in basic critical organs and systems, depending on the nature and dose of radiation effect. It includes some special methods of study of the function of the liver, muscles, hematogenesis, overall adaptive syndrome, reception indicators, motor system and hemodynamics. In addition to purely practical purposes, improvement of diagnostics and determination of rational volume and courses of observation, this will facilitate progress in the general theory of radiobiology in respect to its most complex division, that concerning man /65-68/.

b) More extensive and more purposeful study of the significance of the initial background of activity of non-specific systems which adapt the organism to the external environment and also the state of

critical organs in subsequent reaction to radiation -- as a scientific basis of occupational selection and non-specific prophylaxis of radiation injuries and the remote after-effects of radiation.

c) Rationalization and determination of the volume of effective dosimetric control of radiation safety of persons of different occupations.

d) Study of the most unfavorable sections and forms of work capable of being potential sources of high doses of radiation and the development of all regulations for conducting effective urgent measures during accidents, including adequate dosimetry, routine diagnosis and necessary assistance to sufferers and also measures for eliminating accidents.

e) Further intensification and approximation of dosimetric characteristics to the biological object, especially to man, for obtaining more adequate clinical-radiometric comparisons which facilitate development of the theory of radiobiology in respect to the effect of radiations on man and also the creation of mathematical models which predict possible injuries and which evaluate quantitatively the effectiveness of different therapeutic-prophylactic agents.

f) A study of possibilities of unification of methods of study and statistical methods of processing information on the effect of radiation under occupational conditions which can provide data in a form suitable for machine processing.

Thereby becomes possible the rapid reception of necessary information for individual sectors of industry and for the country as a whole. This information may provide an authentic scientific base for operative planning and coordination of the work of practical institutions, which are involved in problems of radiation safety of persons working with radiations in different countries. They are, in our opinion, a necessary medical component of a system of measures, intended to give mankind not only economically essential resources in the form of wide use of the energy of ionizing radiations for peaceful purposes but also making its use practically safe.

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REMOVAL OF RADIOACTIVE IODINE FROM GASES

New York City PROCEEDINGS OF THE FOURTH INTERNATIONAL CONFERENCE ON THE PEACEFUL USES OF ATOMIC ENERGY in Russian Vol 11, 1972 pp 399-413

[Article by I. Ye. Nakhutin, N. M. Smirnova, V. M. Makarov, G. A. Loshakov, G. A. Laushkina and V. I. Yaroshinskiy, USSR State Committee for the Use of Atomic Energy, Moscow, Union of Soviet Socialist Republics]

[Text] Introduction

The problem of removing radioactive iodine from gases has been discussed in the literature for many years, and it is still a pressing one. Detection of many iodine compounds present in the gas phase adds more complications.

This problem can be divided into two parts, that are virtually unrelated to one another: 1) Removal of iodine from aggressive gases (particularly from those containing nitrogen oxides); 2) removal of iodine from air and other nonaggressive gases.

In both cases, one has to take into consideration the origin of the gases to be treated, since their chemical composition, specific activity, etc., are related to their origin. We submit below the results of some studies on the behavior of iodine in aqueous and nitrate solutions, and about the compounds that iodine forms.

1. Behavior of Iodine in Aqueous and Nitrate Solutions

When nuclear fuel is dissolved, 20 to 80% of the radioiodine remains in the aqueous phase [1, 2]. The amount of radioiodine remaining in the solution depends on the type of fuel, mode of dissolution, equipment used in the process and total iodine content in the nuclear fuel. In order to determine the patterns of distribution of radioiodine in the solution and gas phase, a thermodynamic estimate was made of levels of iodine in different forms, as related to its overall concentration in aqueous solution (with consideration of dissociation of molecular iodine, forming: I^-, I_3^-, I^+, HIO, IO_3 and IO_3^-).

Studies of the kinetics of blowing iodine out of aqueous solutions (Figure 1) revealed that the removal of molecular iodine (I_2) with bubbling air is a first order reaction, while removal of the part of iodine present in the original solution in ionic form (when there are no redox agents) can be described satisfactorily by an equation for a second order reaction:

$$Q_\tau = Q_0^{I_2} e^{-\alpha\gamma\tau} + \frac{Q_0^{ion}}{1 + \frac{1}{2} K \cdot Q_0^{ion} \tau} \qquad (1)$$

where Q_τ is the concentration of iodine in solution at time τ, $Q_0^{I_2}$ is the initial concentration of molecular iodine in the solution, α is the coefficient of distribution of molecular iodine (I_2) in the gas and liquid phases at given temperature, γ is the coefficient that refers to intensity of bubbling and degree of equilibrium between the gas and liquid, Q_0^{ion} is the initial concentration of ionic forms of iodine in the solution, and K is the constant of rate of association of ionic forms, which equals 0.661 $\ell \cdot$g-equiv$^{-1} \cdot$min^{-1} at 80°C.

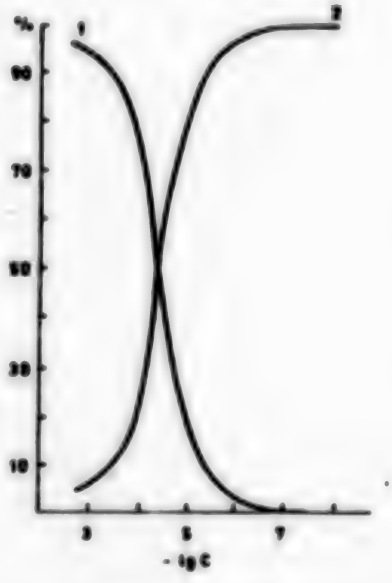

Figure 1.
Change in molecular (1) and ionic (2) forms of iodine content as a function of total concentration of iodine in aqueous solution

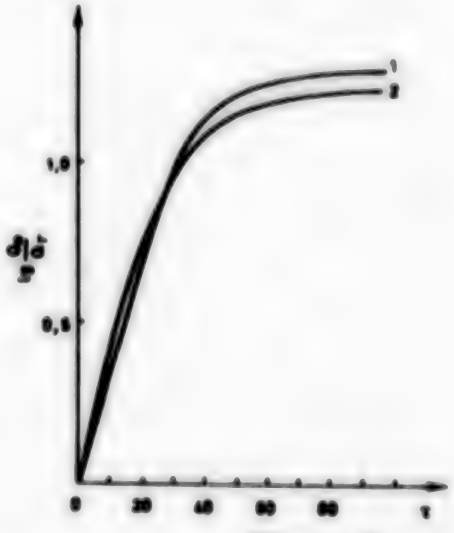

Figure 2.
Kinetics of blow-off of iodine from aqueous solution
1) experimental curve
2) theoretical curve
$Q_0 = Q_0^{I_2} + Q_0^{ion}$ --total initial concentration of iodine
Q_τ) iodine concentration at time τ

Figure 2 illustrates the experimental and theoretical curves for blow-off of iodine from aqueous solution at 80°C and initial total concentration of iodine of $1.12 \cdot 10^{-3}$ g-eq/ℓ.

A satisfactory conformity of estimated and experimental data was obtained as well for blowing iodine out of nitrous oxide solutions. However, the amount of radioiodine remaining in solution after dissolution of nuclear fuel was considerably greater than estimated. Evidently, this result is attributable to occurrence of chemical reactions of iodine during dissolution of nuclear fuel, which were not taken into consideration in the estimates.

The assumption has been made in the literature, to interpret the behavior of radioiodine, that there may be formation of iodine alkyls (in particlar CH_3I), presence in appreciable quantities of a number of unstable compounds such as HIO, HIO_2, INO, INO_2 and INO_3, or accumulation in an oxidative medium of iodine compounds with a high degree of oxidation of IO_3^- or IO_4^- [3-8].

However, none of the mentioned works submitted convincing evidence of the presence of significant amounts of the above-mentioned compounds in the solutions.

In virtually all types of nuclear fuels, there is a carbonitride phase [9-11]. We conducted experiments to determine removal of iodine from solution during dissolution of various materials containing a carbonitride phase, in nitric acid with addition of I_2.

The experiments were conducted with soft iron, tool steel, pig iron, metal uranium and uranium carbonitrides differing in composition. It was established that volatility of iodine decreased and levels thereof in condensates increased with increase in carbonitride phase in the material being dissolved. By means of repeated distillation, a clear, crystalline, readily volatilized iodine compound was isolated from condensates with high iodine content. The results of chemical analysis and studies of infrared spectra revealed that the isolated compound was cyanogen iodide, ICN.

Table 1 lists some data on yield of ICN upon dissolution of various materials in nitric acid containing an excess of I_2.

On the basis of the results of this study, it was established that radioiodine may be present in solutions of nuclear fuel in the form of ICN, I_2 and products of dissociation thereof.

Of the products of dissociation in nitrate media, HIO_3 is the most significant.

During extraction, because of the high solubility of molecular iodine in diluent and formation of a tributyl phosphate—I_2 complex [12] (analogous

complexes are also formed with amines), it changes almost entirely into the organic phase. It is known that I_2 interacts with unsaturated hydrocarbons, forming iodinated hydrocarbons [13]. In the presence of electron acceptors or under the influence of light or radiation, I_2 also interacts with saturated and aromatic hydrocarbons [14-16]. We studied the kinetics of formation of a crude [technical-grade] mixture of saturated hydrocarbons (C_9-C_{15}) in a two-phase system, in the presence of iodine solutions in nitric acid. The rate of iodination can be described by the following equation:

$$V = K \cdot c_{I_2}^{1/2} \cdot a_{HNO_3}^{1/2} \qquad (2)$$

where c_{I_2} is the concentration of iodine in the organic phase, a_{HNO_3} is activity of undissociated HNO_3 in the aqueous phase, and K is the constant of the reaction rate, which equals $1.75 \cdot 10^{-3}$ ℓ/min^{-1} at 20°C.

Table 1. Yield of ICN during dissolution of materials in HNO_3 containing an excess of I_2

Material dissolved	N and C content, g-atom/g-atom metal		ICN yield g-mole/g-atom metal
	N	C	
Soft iron	not analyzed	$4.15 \cdot 10^{-3}$	$6.2 \cdot 10^{-4}$
Tool steel	not analyzed	$6.74 \cdot 10^{-2}$	$5 \cdot 10^{-3}$
Uranium metal, sample No 1	$\sim 8.5 \cdot 10^{-4}$	$1.33 \cdot 10^{-2}$	$2.8 \cdot 10^{-4}$
Uranium metal, sample No 2	$\sim 9 \cdot 10^{-4}$	$3.23 \cdot 10^{-2}$	$5.6 \cdot 10^{-4}$
Uranium carbonitride $UC_{0.62}N_{0.38}$	0.38	0,62	$2.88 \cdot 10^{-2}$

In the presence of TBP [tributyl phosphate] and high-power radiation fields, iodination increases. To reduce accumulation of iodine alkyls in mixture of TBP with diluent, I_2 must be removed from aqueous solutions of nuclear fuel that are to be submitted to extraction.

Iodic acid HIO forms monosolvate with TBP, and the constant of extraction from nitrate solutions is 1.8 ℓ/mole at 20°C.

Cyanogen iodide ICN forms the complex TBP·ICN with TBP, and the extraction constant is 35 ℓ/mole at 20°C. Cyanogen iodide and the complex thereof with TBP are dissociated in alkaline media.

2. Methods of Removing Iodine

2.1. A paper was delivered at one of the conferences [17] pertaining to the use of iodine columns with $AgNO_3$ to trap iodine discharged upon processing of irradiated fuel, in which flaws were reported in the operation of such columns, related to solubility of $AgNO_3$ in water and its fusibility. In 1964, we developed columns for isotope exchange with $Ag^{127}I$, applied to the packing of the column. The advantage of such columns is that AgI is virtually insoluble. Such columns operate on the principle of "permanent" columns, since there is radioactive decay of iodine in silver iodide, along with absorption of radioactive iodine by silver iodide.

We previously derived equations for "permanent" columns for the case of gas-adsorption chromatography [18]. The equations for isotope exchange columns are analogous. According to these equations, there is stationary distribution of concentrations in the column, so that there is exponential decline of concentration of radioactive iodine toward the outlet from the column. The exponent depends on the velocity of gas in the column and rate of mass exchange between gas and solid AgI.

$$c(x) = c_0 \exp\left[-\frac{\lambda \cdot \varphi}{V} \cdot x\right] \qquad (3)$$

where c(x) is the concentration of radioactive iodine at distance "x" from the entrance into the column, c_0 is the concentration of radioactive iodine at entrance into column, λ is the constant of radioactive isotope decay and ϕ is the function describing conditions of mass exchange and radioactive decay in the column; ϕ is unrelated to "c" and "X".

If a high enough value is given to the exponent, a good coefficient of purification can be obtained.

In such a column, the moving force is the difference in isotope concentrations of $Ag^{127}I$ and mixture of isotopes of iodine in the gas phase. The process of equalization of concentrations in the periodically operating column is associated with increase in entropy and, in principle, it does not require exogenous operations. However, in the "permanent" column, it is imperative to keep the difference between concentrations constant to assure continuous operation of the column. This is achieved by the process of radioactive decay.

Apparently, there is no obstruction to passage of stable ^{127}I through the column of isotope exchange, with the exception of a small part thereof, which is expended on the reaction of silver formed after decay of ^{131}I in compound $Ag^{131}I$ in the column packing.

A stationary state is established in the column within a certain time. The transitory processes occurring before establishment of the stationary state can be described as follows: first a chromatographic front is formed in the column, which conforms with equations of frontal chromatography of substances. It is assumed that the time of front formation is considerably shorter than the half-life of the substance, and this is valid in almost all cases, with the exception of very short-lived isotopes. As the front advances through the column, the influence of radioactive decay begins to be felt; for this reason the rear part of the front is an exponent, instead of the straight horizontal line of ordinary frontal chromatography (Figure 3).

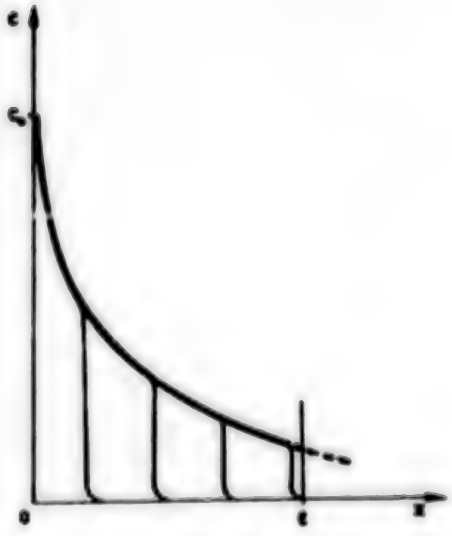

Figure 3.
Advance of front of decaying radioactive substance through column
X) distance from entrance to column
c) concentration of substance

Accordingly the height of the steep part of the front diminishes. One can disregard the influence of radioactive decay on the shape of the steep part of the front. When the advancing steep part of the front reaches the end of the column it is very short, and it is only after it exits from the column that a stationary state is established in the steep part of the front.

The time of establishment of a stationary state is:

$$\tau \sim \frac{Q}{q} \qquad (4)$$

where Q is capacity of column for a given substance and q is amount of substance entering the column per unit time.

The following condition must be satisfied to assure high coefficients of purification:

$$\tau \gg T_{\frac{1}{2}} \qquad (5)$$

where $T_{\frac{1}{2}}$ is the half-life of the radioactive substance.

The solid phase is involved in isotope exchange in the column. Since processes are much slower in the solid phase than the gase phase, one must disperse AgI to assure efficient operation of the column, in order to provide a larger surface and shorter diffusion route in solid particles. Use of sorbent in the pores satisfies these requirements.

Figure 4 illustrates the distribution of radioactive iodine along the isotope exchange column at t = 150°C. The logarithm of concentration in arbitrary units is plotted on the y-axis and distance from entrance into column on the x-axis. The concentration decreases by more than 10,000 times at a distance of 14 cm.

Experiments involving addition of AgI to large-pore sorbent with pore size of 75÷100 Å illustrate the extent of dispersion (Figure 5).

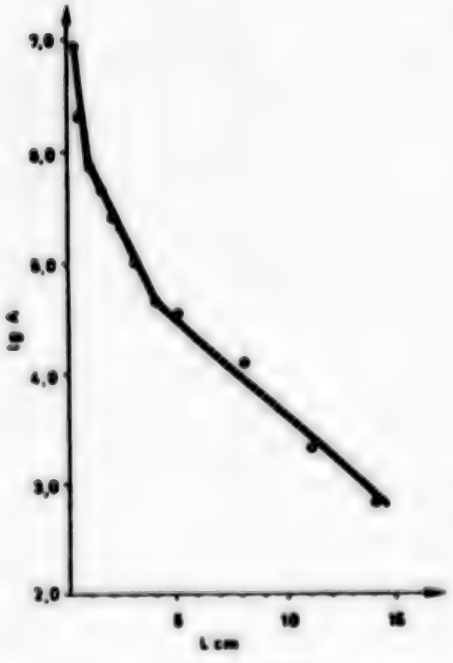

Figure 4.
Distribution of radioactive iodine in isotope exchange column at 150°C. Duration of experiment 180 h; velocity of gas 12 cm/s
L) distance from entrance to column (here and in Figure 5)
A) concentration of radioactive iodine, arbitrary units

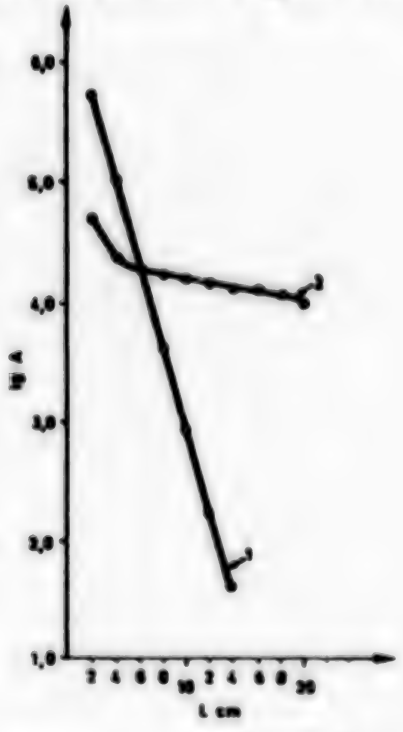

Figure 5.
Distribution of radioactive iodine in isotope exchange columns with small- (curve 1) and large- (2) pore sorbent at 180°C and NO_2 content of ~5%; 10-h experiment; gas velocity 8 cm/s
A) concentration of iodine in arbitrary units

The high melting point of AgI makes it possible to work at rather high temperatures (up to 400°C) (Figure 6). In the temperature range of ~500°C appreciable evaporation of AgI is already observed.

In the presence of nitric oxides dissociation can occur, for example, through the following reaction:

$$2AgI + 2NO_2 + O \rightleftarrows 2AgNO_3 + I_2 \qquad (6)$$

The direction of the reaction is related to partial pressure of NO_2, O_2, I_2 and temperature. The reaction constant as a function of temperature was previously calculated on the basis of thermodynamic data. This reaction is the basis of operation of iodine columns with $AgNO_3$. According to these calculations, one must work at high temperatures to assure stability of AgI. The higher the partial pressure of nitrous oxides and oxygen and the lower the partial pressure of iodine fumes, the higher the working temperature of the column must be. Partial pressure of iodine is determined primarily by the stable isotope of iodine ^{127}I, which is contained in the gas phase.

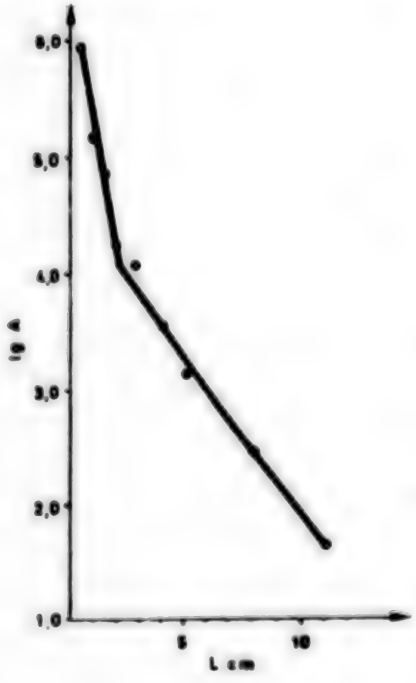

Figure 6.
Distribution of radioactive iodine in isotope exchange column at 400°C. Gas velocity 8 cm/s, time 164 h

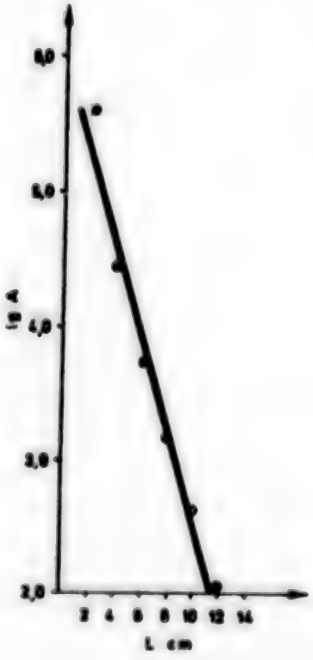

Figure 7.
Distribution of radioactive iodine in isotope exchange column in the presence of ~5% NO_2 at 200°C. Gas velocity 8 cm/s; time 40 h

L) distance from entrance to column
A) concentration of substance in arbitrary units

In actuality, the calculated reaction constants cannot be used for precise quantitative estimates, since $AgNO_3$ and AgI form solid solutions with one another. The partial thermodynamic potentials of solid solutions differ from the thermodynamic potentials of pure substances, and they are not known for the $AgI \cdot AgNO_3$ system. For this reason, the exact minimum required column temperatures as a function of composition of gas mixture cannot be calculated at the present time from the thermodynamic data, and they must be determined from experiments. Obviously, the working temperature must be set at a somewhat higher level than the minimum, in case of possible fluctuations in composition of the gas mixture.

Figure 7 illustrates the distribution of ^{131}I in the isotope exchange column after passage of a gas mixture containing air with addition of 5% NO_2 for 40 h. The logarithm of ^{131}I concentration in arbitrary units is plotted on the y-axis and distance from entrance to column on the x-axis. Column temperature is 200°C and velocity of gas flow 8 cm/s.

We have already mentioned the presence of chemical compounds of iodine in the gas phase, in particular ICN and certain others. The presence of salient points on the logC(x) curve (Figures 4 and 6) is indicative of presence of iodine compounds in the gas flow. Such points appear because function ϕ in equation (3) depends on the nature of the substances involved in isotope exchange and, consequently, different angles of inclination of log C(x) correspond to different substances. However, our findings indicate that radioactive iodine is extracted in isotope exchange columns not only from molecular iodine but from iodine compounds.

The question of term of continuous service of iodine columns merits discussion. Theoretically, "permanent" columns can function as long as desired, since entrance of iodine in the column is equilibrated by its radioactive decay in the solid phase. In practice, however, there may be side reactions that limit the term of column service. One of these reactions is the one with chlorine, which may be present as a trace [microimpurity] in nitric acid and pass from it into the gas phase. As a result, iodine is replaced with chlorine and there is formation of AgCl in the column.

The reaction constants:

$$2AgI + Cl_2 \rightleftharpoons 2AgCl + I_2 \qquad (7)$$

were calculated by us from the thermodynamic data, and they are listed in Table 2.

Table 2 shows that the reaction occurs, even with very low partial pressure of chlorine, lower by many orders of magnitude than the partial pressure of iodine.

Table 2. Constants of equilibrium of the reaction $2AgI + Cl_2 \rightleftarrows 2AgCl + I_2$

Temperature °C	25	100	200	250
K_e	$1,2 \cdot 10^{11}$	$7,5 \cdot 10^{9}$	$1,1 \cdot 10^{7}$	$5,3 \cdot 10^{6}$

Since the admixtures of chlorine are minimal in nitric acid, replacement ["ejection"] is very slow, and the column can function for several years.

2.2. In the case of gas mixtures containing no aggressive gases, activated charcoal is the most effective adsorbent for trapping iodine. Since quite a lot of information has already been published on adsorption of io-iodine in charcoal columns, we shall report only some data that supplement the published material. We submit below some data on equilibrated adsorption of iodine by active charcoal (see Table 3).

Table 3. Adsorption of gas from air at 20°C using activated charcoal No 3 (bulk specific gravity 0.5)

Relative humidity, %	Partial pressure of iodine fumes, torr	Adsorption index
Dry air	10^{-9}	$70 \cdot 10^{6}$
90%	10^{-9}	$14 \cdot 10^{6}$

The dimensionless coefficient [or index] of adsorption "Γ" was calculated using the following formula:

$$\Gamma = \frac{a}{c} \qquad (8)$$

where a is concentration of iodine in charcoal scaled to bulk volume of charcoal and c is concentration of iodine in the gas phase.

Charcoal No 3 does not have the best adsorption index among the existing charcoals. Higher coefficients of adsorption can be obtained with others. The method for determining the adsorption coefficients was selected in such a manner that presence of small amounts of iodine compounds could not affect substantially the results of measurements.

Adsorption equilibrium of iodine is established very slowly. Appreciable changes in equilibrium occurred within 80 h at 20°C. A comparison of the time of establishment of equilibrium at 20°C and 100°C shows that it is reached much faster at high temperatures. This suggests that activated diffusion of iodine in charcoal plays the decisive role, at least at the last stages of the process of establishment of equilibrium.

At low temperatures (-78°C or lower) there is virtually no establishment of adsorption equilibrium between the gas phase and activated charcoal, and we are then dealing with "frozen" nonequilibrated states. As a result of very slow internal diffusion, only the most accessible part of the surface is involved in adsorption. At low temperatures, the apparent coefficient of adsorption decreases with decrease in temperature.

The coefficients of adsorption of iodine are significantly lower on silica gel and zeolites than activated charcoal. For example, with 5 Å zeolite, the coefficient of adsorption is of the order of $\sim 10^4$; however, it is difficult to determine whether this is a real equilibrated value.

The results of experiments dealing with dynamic adsorption of radioactive iodine on activated charcoal were reported by us previously [19]. The theory of adsorption columns, in which dynamic sorption is combined with radioactive decay, was expounded in [18].

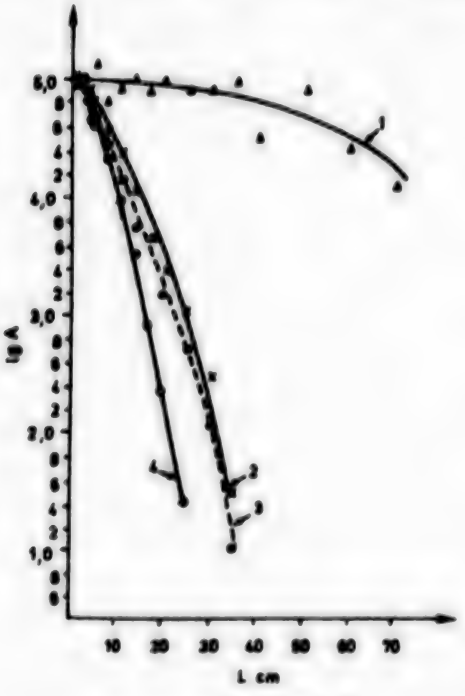

Figure 8.
Distribution of ^{131}I during passage of radioactive methyl iodide through column at 50°C with gas velocity of 30 cm/s; duration of experiment, 25 h
1) charcoal without impregnation
2) CuI-impregnated charcoal
3) PbI_2-impregnated charcoal
4) AgI-impregnated charcoal

Figure 9.
Distribution of ^{131}I during passage of radioactive methyl iodide through column at 50°C; gas velocity 30 cm/s; time 232 h
1) AgI-impregnated charcoal
2) I_2-impregnated charcoal
3) PbI_2-impregnated charcoal

In both figures: L) distance from entrance to column
A) concentration of radioactive iodine

The data in the literature, as well as our experiments, indicate that methyl iodide and other alkyl iodides are present in air, along with molecular iodine. In charcoal columns, these compounds are sorbed only over a rather considerable length; however, it would be desirable to obtain higher purification coefficients with columns that are not overly long.

For this reason, we conducted experiments to test charcoals impregnated with various substances that undergo isotopic exchange with methyl iodide. In addition, these substances must be minimally soluble in water in order to assure operation of the columns after moisture gets into them.

These columns are analogous to the above-described isotope exchange columns with respect to principle of operation. Figures 8 and 9 illustrate the results of experiments with charcoal No 4 impregnated with AgI, PbI_2, CuI, I_2, as well as nonimpregnated charcoal. A mixture of air and radioactive methyl iodide was passed through columns, each of which was equipped with one of the above charcoals. The distribution of concentrations in the columns indicates that the coefficients of purification of methyl iodide are higher by several orders of magnitude for impregnated charcoal than nonimpregnated and, consequently, the effects of purification observed in the columns are related to isotopic exchange between metal iodides and methyl iodide.

In analogous experiments, high purification coefficients persisted at 50°C and partial pressure of water vapor of ~15 torr. Long-term experiments with columns of impregnated charcoal were conducted to establish stationary distribution of concentrations of radioactive iodine in the columns. The experiments lasted 32 days, with overnight breaks. Total work time was 232 h. Figure 9 illustrates the results of these experiments. All of the columns yielded high purification indices. Activity was somewhat lower in the first section of the column impregnated with molecular iodine than in the second. This should probably be attributed to desorption of I_2 from charcoal, which began under the influence of air. The column with PbI_2 behaved somewhat better than the others.

In practice, one has to extract radioactive iodine from gas mixtures, in which methyl iodide constitutes a minor addition to molecular iodine. As a result of poor adsorption of methyl iodide, this addition limits the possibilities of obtaining high purification coefficients. Isotopic exchange of methyl iodide on impregnated charcoal removes this limitation.

In the literature there have already been reports of impregnation of charcoal with potassium iodide and molecular iodine; however, in a number of cases, application on charcoal of compounds that are not soluble in water, such as PbI_2, AgI and CuI, will have advantages.

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RADIOACTIVITY OF OCEAN WATERS AND THE BEHAVIOR OF CERTAIN FISSION PRODUCTS IN THE OCEAN

New York City PROCEEDINGS OF THE FOURTH INTERNATIONAL CONFERENCE ON THE PEACEFUL USES OF ATOMIC ENERGY in Russian Vol 11, 1972 pp 609-630

[Article by D. M. Vdovenko, A. G. Kolesnikov, V. I. Spitsyn, R. N. Bernovskaya, L. I. Gedeonov, V. V. Gromov, L. M. Ivanova, B. A. Nelepo, V. N. Tikhomirov and A. G. Trusov, State Committee on the Peaceful Uses of Atomic Energy in the USSR, Moscow]

[Text] All uses of atomic energy cause contamination of the environment by one or another radioactive products, for the greater part of which the ocean is the ultimate reservoir. The strontium-90 and cesium-137 content in regions of greatest importance in this regard were determined over the period of 1965-1970 to provide the current characteristics of artificial radioactivity in the oceans. The mean strontium-90 and cesium-137 concentration in the surface water was found for each investigated region and its minimum and maximum values were also determined. Data were also obtained on time variation of the concentration. It decreased in the Pacific Ocean and remained at an approximately constant level in the Atlantic Ocean during the period 1965-1970, usually comprising from 15 to 50 disintegrations per minute per 100 liters at latitudes of 70°N and from 30 to 150 disintegrations per minute per 100 liters at latitudes of 40°N. Resorption of previously observed areas of high concentration of radioactivity in the Pacific Ocean was investigated. Locations with increased radioactive contamination were clearly shown to be the result of radioactive waste disposal. Deep-sea samples were taken from the oceans and the strontium-90 and cesium-137 content in the water was estimated. Approximate agreement of this estimate with data from observations of radioactive fallout was noted. Deep currents of water with increased strontium-90 concentration entering the ocean due to waste disposal were found. The behavior of cerium, yttrium and niobium isotopes in the ocean was

studied. The specific effect of sea water on the state of radionuclides was established. Specifically, yttrium-91 is subjected to a significant effect of natural ligands and is in the truly dissolved state in the thermal barrier; approximately 60 percent of the element goes into suspension in other water samples. The biogenic and abiogenic processes of extracting radionuclides from sea water were considered. Abiogenic uptake and fixing of radionuclides by suspension depends on the properties of the substance suspended in the ocean. It is shown that iron, as a biogenic element, contributes to introduction of cerium-144, yttrium-91 and niobium-95 into plankton organisms, that is, into the active biological cycle. The absorption of these nuclides by bottom sediments from sea water in the Indian and Pacific Oceans was studied.

Radioactive waste disposal in the water environment is becoming more massive in nature in the modern world. The role of this process in radioactive contamination of the ocean has become comparable in some locations to the role of global radioactive fallout. The bibliography on radioactive contamination of the ocean is rather fully represented in [1, 2]. Among new publications, [3] and [4] are especially interesting. The latest results of investigations, the previous stages of which were described previously [5, 7], are outlined in this report.

Global radioactive contamination of the ocean was studied mainly from data of observing the concentration of long-lived radionuclides: Sr-90 and Cs-137 [8, 9]. The results of our recent measurements are presented. The behavior of shorter-lived isotopes, including Ce-144, Y-91, Nb-95 and Mo-99 (Tc-99), must be known when studying radioactive contamination occurring due to radioactive waste disposal. The results of the first stages of studying the behavior of these radionuclides in sea water are reported below.

The Sr-90 and Cs-137 content in surface waters at 92 locations of the Pacific Ocean at the end of 1966 and the beginning of 1967 and also at some points of the Atlantic Ocean in 1967 is presented in Table 1. All the data of Table 1 are shown on the charts of Figure 1 and Figure 2. The distribution profiles of Sr-90 and Cs-137 by depth are presented in Table 2. Reliable results were found at all the investigated locations to a depth of 500 meters. The concentration was frequently below the sensitivity of the method at greater depths. Nevertheless, locations were found where the Sr-90 and Cs-137 concentration, even at a depth of 1,000 m and sometimes greater, was above the limit of sensitivity and was determined with sufficient reliability.

The Sr-90 and Cs-137 content in the layer of water to a depth of 500 meters and at some locations to a depth of 1,000 meters was estimated at many points from data on the depth distribution of the concentration. The

results are presented in Table 3. The derived estimate of the content is shown on the chart on Figure 3 and Figure 4. Some characteristics of radioactive contamination of waters by individual regions of the oceans are noted.

The Sr-90 concentration was 71 disintegrations per minute per 100 liters of water in November 1966 and the Cs-137 concentration was 79 disintegrations per minute per 100 liters at one point in the Sea of Japan. Comparison of the figures to data for the Mediterranean and Black Seas [5, 6] shows that the concentration of these radionuclides is higher in all closed seas than in the open ocean at the same latitude. Global radioactive fallout from 1962 through 1966 decreased [10], but the Sr-90 and Cs-137 concentration in these seas hardly diminished. The Sr-90 and Cs-137 content in the layer of water from the surface to a depth of 500 meters in the Sea of Japan was 95 and 107 millicuries per square kilometer and that to a depth of 1,000 meters was 135 and 162 millicuries per square kilometer, respectively. According to data of the United Nations Scientific Committee on the Effects of Atomic Radiation, found from observations on land stations, one should expect 60-80 millicuries per square centimeter of Sr-90 in this region [11]. Excess Sr-90 is present.

The waters of the northwestern Pacific Ocean were contaminated due to nuclear arms testing in the Marshall Islands. A region of extreme radioactive contamination was observed here in 1954 and 1955. The position was quite different in the fall of 1966. The results were compared to data of past years [12, 13] in Figure 5, in which data for the Atlantic are also presented [5, 6]. The Sr-90 content in the surface water of this region decreased significantly during recent years and comprised an average of 30 disintegrations per minute per 100 liters in the fall of 1966. The Cs-137 concentration was an average of 49 disintegrations per minute per 100 liters. The zone of increased concentration in the region of Bikini-Eniwetok had essentially disappeared. The Sr-90 and Cs-137 content in the upper 500 meters of water no longer differed from that in other regions of the northwestern Pacific Ocean. True, the upper layer of 1,000 meters contains 115 millicuries per square kilometer of Cs-137 over the Marianis Trench. The concentration in the Marianis Trench at great depths is below the range of sensitivity. Samples were taken here to a depth of 8,000 meters.

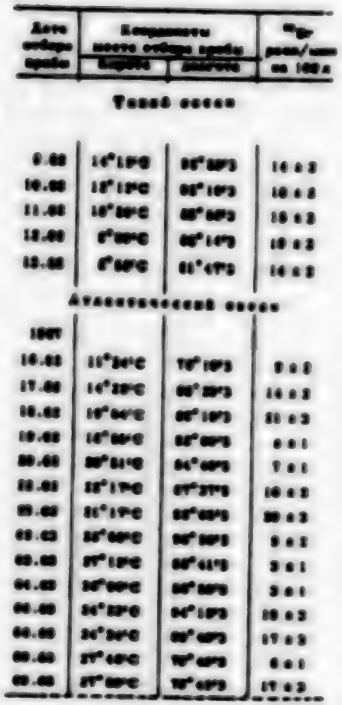
The Sr-90 and Cs-137 concentration in the southwestern Pacific Ocean was less in the surface waters than in the northwestern part. Comparison of the average figures is given in Table 4. Data for the Atlantic Ocean are also presented in this table [6]. The ratio was maintained from year to year. Detectable amounts of Sr-90 and Cs-137 existed only in the upper layer of the water above the deepwater Kermadec Trench. Deep waters to 9,000 meters were pure, although they were not quite stagnant according to preliminary hydrological data [16] and may be supplemented due to dropping of the water mass in other regions.

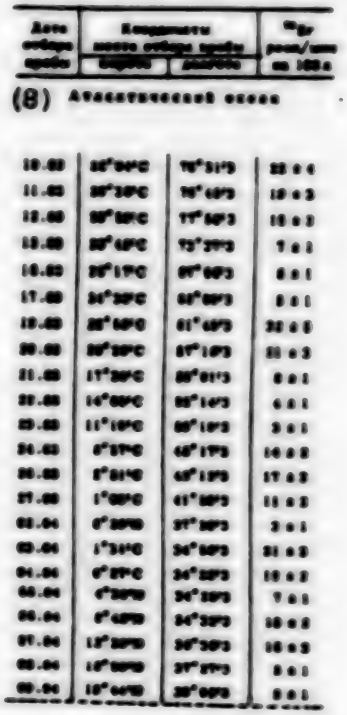
Table 1. Sr-90 and Cs-137 Content in the Surface Water of the Oceans

(1) Дата отбора пробы	(2) Координаты места отбора пробы: широта	долгота	(5) Концентрация расп/мин · 100 л: ^{90}Sr	^{137}Cs	(6) Отнош. $^{137}Cs/^{90}Sr$
	(3)	(4)	Тихий океан (7)		
1966					
1.11	42°19°С	134°30°В	71 ± 11	79 ± 11	1,1
5.11	40°00°С	142°15°В	64 ± 7	66 ± 9	1,0
8.11	36°00°С	142°41°В	68 ± 7	78 ± 11	1,1
8.11	30°04°С	144°07°В	43 ± 7	38 ± 10	1,0
9.11	28°10°С	145°17°В	36 ± 5	78 ± 8	1,0
10.11	23°30°С	145°40°В	43 ± 6	51 ± 11	1,2
11.11	19°30°С	147°28°В	49 ± 7	56 ± 10	1,1
13.11	13°00°С	147°00°В	43 ± 7	58 ± 7	1,3
14.11	13°40°С	148°48°В	18 ± 3	33 ± 3	1,8
15.11	13°22°С	152°24°В	23 ± 4	44 ± 6	1,9
16.11	12°58°С	154°47°В	21 ± 3	41 ± 6	1,9
17.11	12°35°С	158°72°В	22 ± 4	42 ± 6	1,9
18.11	11°42°С	161°30°В	22 ± 4	38 ± 6	1,7
19.11	12°00°С	162°10°В	30 ± 6	56 ± 14	1,8
20.11	12°09°С	164°48°В	21 ± 3	40 ± 9	1,9
21.11	12°01°С	167°13°В	14 ± 3	24 ± 6	1,7
22.11	10°33°С	170°28°В	10 ± 2	15 ± 5	1,5
23.11	8°28°С	171°19°В	18 ± 3	31 ± 9	1,8
24.11	6°43°С	169°49°В	36 ± 6	42 ± 11	1,1
25.11	6°00°С	167°11°В	21 ± 5	38 ± 14	1,8
26.11	3°32°Ю	165°00°В	25 ± 3	37 ± 11	1,5
27.11	1°00°Ю	162°50°В	27 ± 4	33 ± 11	1,2
29.11	15°30°Ю	156°43°В	9 ± 2	10	–
30.11	16°04°Ю	151°30°В	11 ± 2	11 ± 2	1,0
1.12	21°10°Ю	151°22°В	13 ± 3	21 ± 6	1,7
2.12	25°10°Ю	151°10°В	10 ± 2	–	–
3.12	28°30°Ю	154°48°В	17 ± 3	28 ± 9	1,6
4.12	32°03°Ю	152°20°В	22 ± 4	32 ± 13	1,4
5.12	35°03°Ю	152°30°В	26 ± 4	30 ± 12	1,1
10.12	36°48°С	153°23°В	27 ± 4	37 ± 13	1,4
11.12	33°20°Ю	156°30°В	11 ± 2	13 ± 7	1,2
12.12	33°01°Ю	161°04°В	10 ± 2	16 ± 7	1,6
13.12	33°41°Ю	169°19°В	8 ± 2	–	–
14.12	32°30°Ю	174°58°В	9 ± 2	–	–
15.12	32°09°Ю	178°19°В	10 ± 2	–	–
16.12	32°03°С	177°20°З	9 ± 2	–	–
16.12	32°03°Ю	177°20°З	11 ± 2	10	–
18.12	30°43°Ю	176°04°З	9 ± 2	16 ± 7	1,7
19.12	28°07°Ю	174°39°З	9 ± 2	10	–
21.12	26°13°Ю	173°10°З	30 ± 5	–	–
23.12	19°30°Ю	166°03°З	85 ± 8	–	–
25.12	18°47°Ю	160°34°З	130 ± 25	–	–
26.12	17°07°Ю	157°05°З	85 ± 6	28 ± 9	1,0
27.12	17°20°Ю	152°40°З	19 ± 3	–	–
30.12	18°03°Ю	148°43°З	31 ± 5	–	–
30.12	18°43°Ю	144°04°З	13 ± 3	22 ± 8	1,7
31.12	18°22°Ю	142°32°З	10 ± 2	17 ± 7	1,7
1967					
1.01	20°14°Ю	140°24°З	13 ± 3	18 ± 9	1,3
3.01	19°30°Ю	136°27°З	13 ± 3	15 ± 6	1,1
4.01	18°09°Ю	137°30°З	9 ± 2	16 ± 6	1,7
5.01	11°40°Ю	136°43°З	7 ± 1	13 ± 7	1,8
6.01	9°48°Ю	135°28°З	10 ± 2	–	–
7.01	8°47°Ю	135°43°З	8 ± 1	–	–
8.01	1°12°Ю	134°08°З	9 ± 2	–	–
9.01	0°34°С	134°30°З	10 ± 2	–	–
10.01	4°38°С	133°18°З	10 ± 2	–	–
11.01	8°07°С	132°07°З	12 ± 2	–	–
12.01	12°14°С	131°04°З	17 ± 3	–	–
13.01	15°32°С	130°20°З	17 ± 3	–	–
14.01	20°10°С	128°04°З	24 ± 4	–	–
15.01	23°41°С	127°42°З	34 ± 5	–	–
16.01	26°08°С	126°20°З	62 ± 10	–	–
17.01	33°10°С	125°13°З	68 ± 10	–	–
24.01	36°21°С	125°07°З	29 ± 6	–	–
25.01	41°15°С	126°20°З	78 ± 25	–	–
26.01	43°32°С	127°00°З	38 ± 10	106 ± 12	2,8
26.01	41°10°С	126°00°З	66 ± 10	118 ± 13	1,8
26.01	44°31°С	125°37°З	80 ± 14	122 ± 14	1,5
27.01	45°13°С	125°07°З	68 ± 10	110 ± 13	1,6
27.01	45°32°С	124°51°З	64 ± 10	88 ± 10	1,4
27.01	46°14°С	124°43°З	60 ± 10	89 ± 10	1,5
28.01	46°38°С	124°28°З	55 ± 8	78 ± 10	1,4
28.01	46°52°С	124°28°З	65 ± 7	73 ± 10	1,0
29.01	46°41°С	125°48°З	80 ± 10	100 ± 11	1,3
29.01	46°32°С	126°28°З	75 ± 11	107 ± 12	1,4
29.01	46°37°С	126°08°З	80 ± 8	150 ± 11	2,0
29.01	44°39°С	125°58°З	51 ± 7	90 ± 12	1,7
30.01	43°10°С	125°18°З	62 ± 9	105 ± 13	1,7
31.01	39°10°С	123°58°З	39 ± 6	–	–
1.02	35°07°С	122°43°З	62 ± 9	84 ± 9	1,3
2.02	31°14°С	120°15°З	46 ± 7	–	–
3.02	26°06°С	116°46°З	97 ± 9	110 ± 13	2,0
4.02	25°01°С	113°31°З	47 ± 7	–	–
5.02	23°13°С	110°16°З	29 ± 6	–	–
6.02	20°33°С	107°41°З	31 ± 8	–	–
7.02	18°19°С	103°58°З	16 ± 3	–	–
8.02	16°21°С	99°39°З	25 ± 4	–	–

[Table continued on following page]

Table 1 (Continued).





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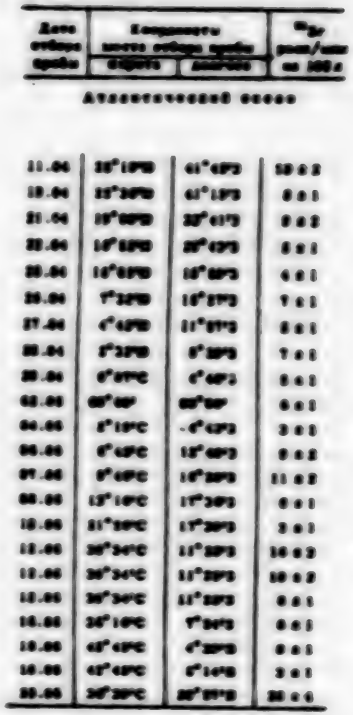

Key:

1. Date of taking sample
2. Coordinates of taking sample
3. Latitude
4. Longitude
5. Concentration, disintegrations per minute per 100 liters
6. Ratio
7. Pacific Ocean
8. Atlantic Ocean

Table 2. Sr-90 and Cs-137 Content in the Deep Waters of the Oceans

(1) Дата отбора пробы	(2) Координаты	(3) Глубина океана, м	(4) Глубина горизонта отбора пробы, м	(5) Концентрация: пкюри/литр ^{90}Sr	(5) Концентрация: пкюри/литр 100 х ^{137}Cs	(6) Отношение $^{137}Cs/^{90}Sr$
			Тихий океан (7)			
1966						
1.11	41°13'С 134°32'З	3700	0	71 ± 11	79 ± 11	1,1
			200	60 ± 6	66 ± 6	1,1
			900	26 ± 5	27 ± 7	1,0
			1800	7 ± 3	10 ± 5	1,4
6.11	36°00'С 142°41'В	6412	0	65 ± 7	78 ± 11	1,2
			500	16 ± 3	–	–
			1000	8 ± 2	10	–
			3000	5	10	–
10.11	22°30'С 148°48'В	5973	0	43 ± 7	61 ± 11	1,4
			500	19 ± 3	27 ± 10	1,4
			1000	15 ± 3	21 ± 6	1,4
			3000	6 ± 1	11 ± 6	1,8
			7500	10 ± 2	17 ± 6	1,7
11.11	19°30'С 147°39'В	6830	0	49 ± 7	58 ± 10	1,2
			200	37 ± 6	48 ± 8	1,3
			500	11 ± 3	10	–
			1000	11 ± 3	10	–
			2000	5	10	–
			3000	5	–	–
			6000	5	10	–
12.11	13°00'С 147°00'В	5265	0	43 ± 7	52 ± 7	1,2
			150	30 ± 6	66 ± 9	1,7
			200	18 ± 4	23 ± 5	1,3
			500	9 ± 3	10	–
			1800	–	–	–
			4000	5	10	–
18.11	11°43'С 161°30'В	1940	0	22 ± 4	42 ± 6	1,9
			150	22 ± 4	35 ± 3	1,5
			5500	10 ± 2	–	–
			1800	7 ± 4	–	–

Дата отбора пробы	Координаты	Глубина океана, м	Глубина горизонта отбора пробы, м	Концентрация: пкюри/литр ^{90}Sr	Концентрация: пкюри/литр 100 х ^{137}Cs	Отношение $^{137}Cs/^{90}Sr$
			Тихий океан			
19.11			2000	5 ± 2	10	–
			3000	5 ± 2	10	–
			5200	5	10	–
			8000	5	10	–
21.11	20°13'С 173°10'З	5527	0	30 ± 5	–	–
			800	16 ± 3	–	–
			1000	5	10	–
			2000	5 ± 2	–	–
			3000	5	10	–
			4000	5	10	–
			5000	5	10	–
26.11	17°07'С 150°02'З	4213	0	26 ± 4	26 ± 5	1,0
			150	18 ± 3	19 ± 4	1,0
			2000	16 ± 3	20 ± 5	1,3
30.11	10°43'С 140°34'З	4356	0	13 ± 2	23 ± 5	1,7
			500	10 ± 2	15 ± 5	1,5
30.12	19°25'С 142°32'З	4418	0	10 ± 2	17 ± 7	1,7
			800	5	10	–
			1000	5	10	–
			2000	5	–	–
			3000	5	10	–
1967						
1.01	20°14'С 140°34'З	4985	0	13 ± 2	18 ± 5	1,5
			500	5	10	–
			1000	5	10	–
			2000	5	10	–
			4000	5	10	–
2.01	18°30'С 138°27'З	5457	0	13 ± 2	15 ± 5	1,2
			200	7 ± 2	13 ± 5	1,7
			800	7 ± 2	–	–
			1000	5	10	–
			2000	–	–	–

[Table continued on following page]

Table 2 (Continued).

– 10.11	12°06'S 142°10'E	2880	0 500 1000 2020	30 ± 6 15 ± 3 11 ± 5 17 ± 2	46 ± 14 20 ± 7 14 ± 6 29 ± 9	1,0 1,3 1,3 1,7
20.11	12°06'S 144°48'E	1867	0 250 500 1000 1500	21 ± 3 9 ± 3 9 ± 2 6 ± 1 5 ± 3	40 ± 9 10 ± 7 17 ± 6 10 ± 5 10	1,0 1,0 1,0 1,0 –
30.11	0°06'S 167°11'E	4342	0 500 1000 2000 4000	31 ± 6 11 ± 5 7 ± 3 5 5	36 ± 14 17 ± 6 12 ± 6 10 10	1,2 1,0 1,7 – –
30.11	23°30'S 166°42'E	3000	0 500 1000	9 ± 2 5 ± 3 5 ± 1	10 10 10	– – –
3.12	25°10'S 167°10'E	3788	0 500 1000 2000	10 ± 2 10 ± 2 12 ± 3 5 ± 2	– 10 ± 3 12 ± 6 10	– 1,0 1,0 –
12.12	25°01'S 168°04'E	3131	0 500 1000 2000	10 ± 2 5 ± 2 5 5	10 ± 7 – – –	1,0 – – –
19.12	28°03'S 179°30'E	10000	0 500 1000	11 ± 2 7 ± 3 5 ± 2	10 10 10	– – –
19.12	28°03'S 179°30'E	10000	2000 4700 9000	5 5 ± 2 5	10 10 10	– – –
19.12	27°42'S 176°08'E	6881	0 500 7000	9 ± 2 5 5	10 ± 7 10 10	1,7 – –
19.12	27°07'S 174°38'E	10141	0 500 1000	9 ± 2 7 ± 3 5 ± 2	10 – 10	– – –

4.01	10°00'S 137°30'E	4039	0 500 1000 2000 3664	9 ± 2 5 5 5 5	16 ± 6 – 10 10 10	1,7 – – – –
5.01	11°40'S 139°43'E	4178	0 500 1000 2000 3000	7 ± 1 5 5 5 5	13 ± 7 10 10 10 10	1,0 – – – –
8.01	1°12'S 134°58'E	4183	0 500	9 ± 2 5 ± 2	– –	– –
12.01	13°14'S 131°04'E	1887	0 500 1000 2000	17 ± 1 5 5 5	– 10 – 10	– – – –
20.01	42°22'S	2024	0 500 1000	126 ± 18 111 ± 17 21 ± 2	100 ± 12 45 ± 10 –	0,9 0,4 –
26.01	44°10'S 126°04'E	2041	0 507 1000	106 ± 15 68 ± 10 17 ± 3	110 ± 12 – 20 ± 10	1,1 – 1,2
26.01	44°27'S 128°27'E	2981	0 500 1000	90 ± 13 52 ± 8 40 ± 6	123 ± 14 – 20 ± 12	1,2 – 0,9
27.01	45°12'S 128°07'E	1887	0 500 1000	68 ± 10 18 ± 3 8 ± 1	110 ± 12 10 ± 8 14 ± 7	1,0 1,0 1,7
27.01	45°22'S 126°51'E	981	0 500	84 ± 10 8 ± 2	80 ± 10 10	1,0 –
27.01	45°14'S 126°43'E	788	0 250	80 ± 9 41 ± 7	80 ± 10 23 ± 7	1,2 0,6
28.01	46°29'S 124°29'E	603	0 250	55 ± 8 19 ± 3	70 ± 10 10	1,4 –
30.01	46°52'S 124°48'E	178	0 100	45 ± 7 25 ± 5	73 ± 10 60 ± 10	1,0 1,0

[Table continued on following page]

Table 2 (Continued).

Дата отбора пробы	Координаты	Глубина океана, м	Глубина горизонта отбора пробы	Концентрация распад/мин. 100 л ^{90}Sr	Концентрация распад/мин. 100 л ^{137}Cs	Отношение $^{137}Cs/^{90}Sr$
			Тихий океан			
28.01	46°32'С 126°28'З	2878	0	79±11	107±12	1,4
			500	5±1	–	–
			1000	5±1	10	–
9.02	22°12'С 110°19'З	3300	0	38±6	–	–
			500	6	–	–
			1000	5	–	–
			2000	5	–	–
			Атлантический океан (8)			
1967						
18.08	11°26'С 80°19'З	3800	0	9±1	–	–
			500	2±1	–	–
			1000	1±1	–	–
			2000	1±1	–	–
20.08	21°48'С 86°19'З	1480	0	11±2	–	–
			200	7±1	–	–
			500	10±2	–	–
			800	3±1	–	–
02.09	25°00'С 86°30'З	3800	0	9±1	–	–
			500	7±1	–	–
			1000	10±2	–	–
			2000	1±1	–	–
			3000	3±1	–	–
03.09	27°12'С 86°41'З	1800	0	3±1	–	–
			500	3±1	–	–
			1000	4±1	–	–
04.09	28°00'С 86°30'З	3100	0	3±1	–	–
			500	10±3	–	–
			1000	8±1	–	–
			1800	27±4	–	–
08.09	27°30'С 79°48'З	430	0	17±3	–	–
			100	5±1	–	–
			250	4±1	–	–
			300	2±1	–	–
10.09	26°04'С 79°31'З	730	0	22±4	–	–
			300	21±3	–	–
			500	16±3	–	–
			700	1±1	–	–
11.09	25°30'С 78°45'З	700	0	19±3	–	–
			300	9±1	–	–
12.09	25°00'С 77°05'З	800	0	18±2	–	–
			700	3±1	–	–

Дата отбора пробы	Координаты	Глубина океана, м	Глубина горизонта отбора пробы	Концентрация распад/мин. 100 л ^{90}Sr	Концентрация распад/мин. 100 л ^{137}Cs	Отношение $^{137}Cs/^{90}Sr$
			Атлантический океан			
17.03	26°30'С 65°09'З	5800	0	5±1	–	–
			500	4±1	–	–
22.03	14°00'С 52°14'З	4800	0	4±1	–	–
			500	1±1	–	–
			1000	8±1	–	–
			4000	2±1	–	–
27.03	1°00'С 41°58'З	4300	0	11±2	–	–
			500	3±1	–	–
			1000	1±1	–	–
			2000	1±1	–	–
02.04	0°20'Ю 27°38'З	4500	0	3±1	–	–
			500	1±1	–	–
			1000	1±1	–	–
			2000	3±1	–	–
			2500	3±1	–	–
03.04	1°21'С 34°58'З	4000	0	21±3	–	–
			500	6±1	–	–
			1000	4±1	–	–
			2000	2±1	–	–
04.04	0°27'С 34°29'З	4000	0	12±2	–	–
			500	5±1	–	–
			800	8±1	–	–
			1000	4±1	–	–
			3200	1±1	–	–
11.04	25°19'Ю 41°48'З	3400	0	18±2	–	–
			300	36±6	–	–
			1000	4±1	–	–
29.04	0°07'С 6°48'З	5170	0	8±1	–	–
			500	8±1	–	–
			2000	1±1	–	–
			3400	7±1	–	–
08.08	13°10'С 17°30'З	800	0	6±1	–	–
			250	3±1	–	–
			500	5±1	–	–
10.08	42°42'С 4°33'В	1200	0	8±1	–	–
			500	1±1	–	–
			1000	1±1	–	–
10.08	42°45'С 5°14'В	1200	0	3±1	–	–
			500	2±1	–	–
			1000	1±1	–	–

Key:

1. Date of taking sample
2. Coordinates
3. Depth of ocean, meters
4. Depth of sampling level
5. Concentration, disintegrations per minute per 100 liters
6. Ratio
7. Pacific Ocean
8. Atlantic Ocean

Table 3. Sr-90 and Cs-137 Content at Individual Points of the Pacific and Atlantic Oceans in the Water Layer From the Surface to a Depth of 1,000 m

Дата разреза (1)	Координаты разреза (2)		Запас мКи/км² в слое от поверхности до глубины (5)			
			500 м		1000 м	
	широта (3)	долгота (4)	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
Тихий океан (6)						
...6						
1.11	61°19' С	134°32' В	98	117	136	162
6.11	36°00 С	142°41' В	70	-	93	-
10.11	29°30' С	148°40' В	70	135	115	180
11.11	19°30' С	147°29' ВД	73	-	98	-
13.11	13°30' С	147°00' В	50	-	-	-
18.11	11°42' С	161°30' В	63	-	85	-
19.11	12°00' С	162°10' В	55	85	85	135
20.11	12°00' С	164°40' В	35	50	46	80
25.11	0°00' С	167°11' В	60	68	70	100
29.11	13°30' Ю	166°42' В	18	-	30	-
2.12	25°10' Ю	167°10' В	25	-	50	-
12.12	33°01' В	165°00' В	20	-	-	-
16.12	32°00' Ю	177°30' З	-	-	33	-
18.12	33°07' Ю	174°30' З	30	-	38	-
21.12	30°12' Ю	172°10' З	50	-	-	-
30.12	18°45' Ю	144°50' З	30	45	-	-
1967						
2.01	19°50' Ю	130°27' З	18	-	-	-
8.01	01°12' Ю	136°50' З	18	-	-	-
25.01	43°32' С	137°50' З	270	168	450	-
26.01	44°10' С	138°00' З	190	-	350	350
26.01	44°37' С	138°37' З	180	-	270	360

Дата разреза	Координаты разреза		Запас мКи/км² в слое от поверхности до глубины			
			500 м		1000 м	
	широта	долгота	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
Тихий океан						
27.01	45°13' С	135°07' З	87	143	130	190
27.01	45°32' С	136°32' З	80	-	-	-
27.01	46°14' С	136°43' З	94	77	-	-
28.01	46°30' С	136°30' З	85	-	-	-
29.01	46°41' С	135°49' З	90	135	150	-
29.01	46°32' С	136°30' З	90	-	180	-
1967	Атлантический океан (7)					
16.02	11°30' С	60°10' З	50	-	-	-
20.02	21°40' С	64°10' З	20	-	-	-
1.03	23°00' С	60°50' З	18	-	22	-
4.03	25°00' С	64°59' З	21	-	49	-
10.03	28°04' С	70°31' З	44	-	-	-
12.03	29°58' С	77°58' З	24	-	-	-
17.03	34°30' С	65°00' З	10	-	-	-
23.03	16°00' С	52°10' З	5,8	-	17	-
27.03	1°00' С	41°58' З	16	-	-	-
3.04	0°30' Ю	37°30' З	4,8	-	-	-
5.04	1°30' С	34°58' З	20	-	41	-
6.04	0°27' С	34°29' З	21	-	27	-
11.04	25°10' С	61°49' З	60	-	188	-
20.04	0°07' С	4°48' З	15	-	-	-
8.05	12°10' С	17°30' З	10	-	-	-

Key:

1. Date of profile
2. Coordinates of profile
3. Latitude
4. Longitude
5. Content, millicuries per square kilometer in the layer from the surface to a depth of
6. Pacific Ocean
7. Atlantic Ocean

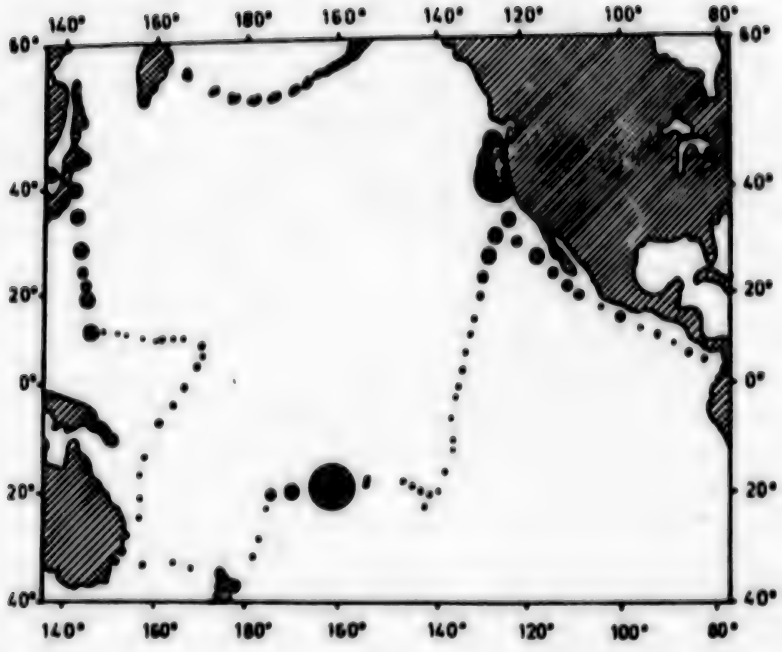

Figure 1. Sr-90 Concentration in the Surface Waters of the Pacific Ocean During 1966-1967 (the diameter of the circles is proportional to the extent of concentration)

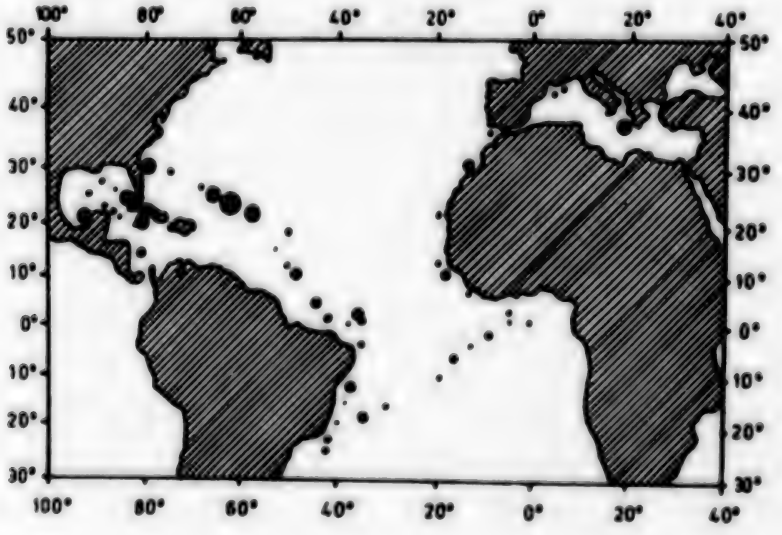

Figure 2. Sr-90 Concentration in the Surface Waters of the Atlantic Ocean in 1967 (the diameter of the circles is proportional to the extent of concentration)

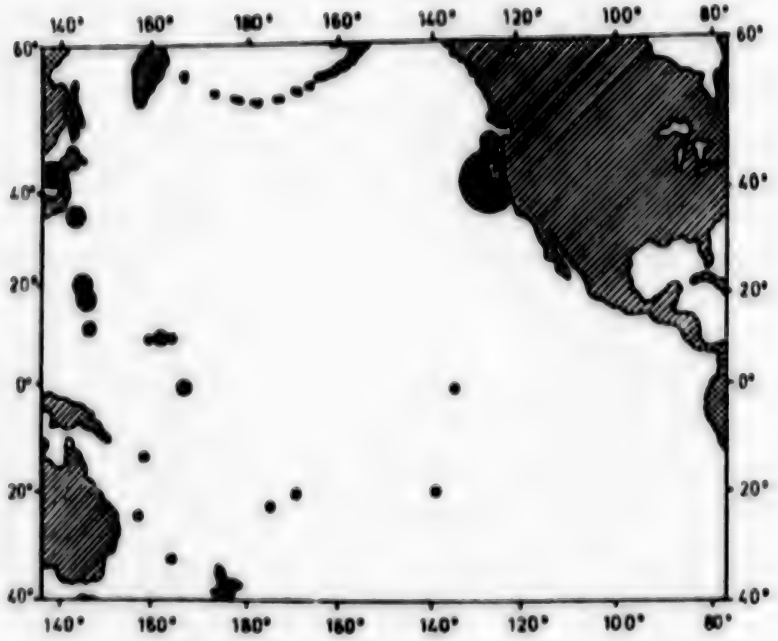

Figure 3. Sr-90 Content in Layer of Water to 500 Meters Per Unit Area of the Pacific Ocean Basin

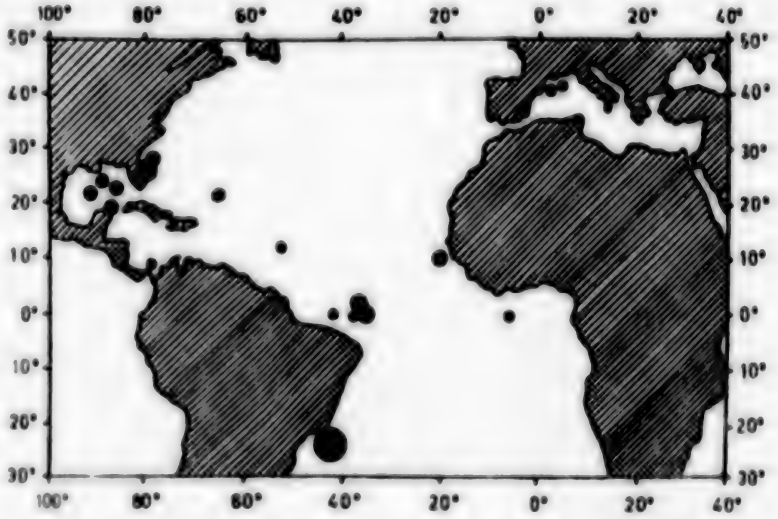

Figure 4. Sr-90 Content in the Water Layer to 500 Meters Per Unit Area of the Atlantic Ocean Basin

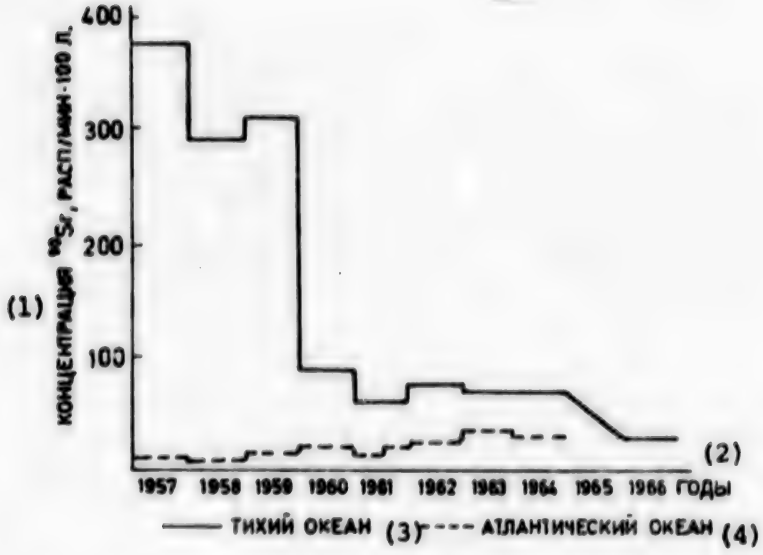

Figure 5. Variation of Sr-90 Concentration in Surface Water of the Northern Pacific Ocean and the Atlantic Ocean During the Period 1957-1966

Key:

1. Sr-90 concentration, disintegrations per minute per 100 liters
2. Years
3. Pacific Ocean
4. Atlantic Ocean

Table 4. Comparison of Mean Sr-90 and Cs-137 Concentration in the Northern and Southern Parts of the Pacific and Atlantic Oceans at Lower Latitudes

(1) Океан	(2) Нуклид	(3) Год	(4) Концентрация расп/мин. на 100 л		(7) Отнош. сев./южн.
			(5) северн.	(6) южн.	
(8) Тихий	(9) Стронций-90	1961 [13,14]	60	30	2,0
	Стронций-90	1966	30	14	2,1
	Цезий-137 (10)		49	24	2,0
(11) Атлантический	Стронций-90	1963 [6]	39	22	1,8
	Цезий-137		77	35	2,2
	Стронций-90	1964 [6]	29	18	1,6
	Цезий-137		50	26	1,9

Key:

1. Ocean
2. Nuclide
3. Year
4. Concentration, disintegrations per minute per 100 liters

[Key continued on following page]

[Key continued from following page]

5. Northern
6. Southern
7. Northern/southern ratio
8. Pacific
9. Strontium-90
10. Cesium-137
11. Atlantic

The mean value of Sr-90 concentration was 57 disintegrations per minute per 100 liters in the narrow latitudinal zone from 17°S to 23°S. A point in the region of the deepwater Tonga Trench with surface water contamination was found; the Sr-90 concentration was 180 disintegrations per minute per 100 liters. High density of radioactive fallout from the atmosphere was observed in this section when samples were taken. This also apparently explains the increased Sr-90 content at the indicated location (18°47'S and 160°35'W). The Sr-90 concentration was below the range of sensitivity of the method at all depths below the water surface above the Tonga Trench. The Sr-90 content in surface waters was not elevated in any way at other points of this zone. True, according to data of [17], the presence of Ce-141, Ce-144, Ru-103, Zr-95 and Nb-95 was found in zooplankton samples at these same locations. The Sr-90 and Cs-137 concentration in surface waters was not as high in the section from the Tuamotu Archipelago to the California coast. The Sr-90 concentration in the southern hemisphere was an average of 17 disintegrations per minute per 100 liters and it was 35 disintegrations per minute per 100 liters in the northern hemisphere from the equator to San Francisco. The Cs-137 content in the region of Polynesia was an average of 18 disintegrations per minute per 100 liters.

Increased Sr-90 and Cs-137 concentration in the region of the Columbia River delta was found in January 1967 at more northerly latitudes. It was possible to compare our results to data obtained by American investigators in 1964 [18]. The mean values in disintegrations per minute per 100 liters in this region were 109 for Sr-90 and 102 for Cs-137 and the ratio of Cs-137/Sr-90 was 0.9 in July 1964 and the values were 78 for Sr-90 and 100 for Cs-137 and the ratio of Cs-137/Sr-90 was 1.3 in January 1967. It was typical for the waters to become much fresher near the mouth of the river in the summer. The river water was characterized by a Cs-137/Sr-90 ratio of approximately 0.3 in the summer. The ocean water had normal salinity (35 percent) and the ratio was approximately 1.8. An intermediate ratio of 1.3 was observed in our case. It is closer to data for the open ocean than to data for river water (70 percent of the water of the open ocean and 30 percent river water); no data were given on the salinity of this ratio. Thus, radioactive contamination in this region was the same in 1967 as in 1964 with lesser contamination of river water. The Sr-90 and Cs-137 concentration in the surface water was significantly higher in absolute value here than at other points of the Pacific Ocean.

Sr-90 content was determined at six points and Cs-137 content was determined at three points in this region in the 100-meter level of water. The values of 100-420 millicuries per square kilometer of Sr-90 and 180-340

millicuries per square kilometer of Cs-137 are completely beyond the range found in the remaining Pacific Ocean basin. Comparison to data on accumulation of fallout Sr-90 [11] shows that global radioactive fallout could produce only approximately 20 percent of the observed content. The content was estimated at approximately 70,000 curies for Sr-90 and approximately 100,000 curies for Cs-137 in the sector of ocean with radius of 435 km, area of approximately 220,000 km^2 and with the center at the mouth of the Columbia River.

The Sr-90 concentration in the Pacific Ocean near the American coast from 35°N to 5°N was approximately the same in 1967 as in the northwestern part of the ocean. It comprised an average of 31 disintegrations per minute per 100 liters for 13 measurements and decreased from north to south.

Summaries of the Sr-90 content in the Atlantic Ocean are presented in [19, 20]. However, the results of measurements only at temperate latitudes are given for 1967. Our investigations of 1967 mainly encompassed the tropical zone of the Atlantic.

The waters of the Caribbean are formed by the northern equatorial current. The mean value of Sr-90 concentration was 11 disintegrations per minute per 100 liters and did not differ from its content in the tropical zone of the ocean. The Sr-90 content in deep waters was below the sensitivity of the method. The Sr-90 concentration in the Yucatan Channel was essentially identical from the surface to 500 meters.

Artificial radioactive products from the mainland apparently enter the Gulf of Mexico. Diffusion and advective transfer result in transformation of water masses then carried away by the gulf stream. An increase of Sr-90 concentration to 18 disintegrations per minute per 100 liters was noted here. The radioactive contamination penetrated to a depth of 1,800 meters at one of the observation points.

Ocean water masses from the north equatorial current move in the western part of the Sargasso Sea. The Sr-90 concentration is within the range of 8-10 disintegrations per minute per 100 liters.

The average Sr-90 concentration was 11 disintegrations per minute per 100 liters in the spring of 1967 in the equatorial zone of the Atlantic Ocean from 5°N to 5°S. No higher concentrations were observed than in the adjacent zones, as was the case in 1963 [6, 20]. The area of abnormally high concentration which was noted in the fall of 1963 also disappeared [6].

There was equalizaton in Sr-90 distribution in the southern part of the tropical zone by 1967; its mean content was 9 disintegrations per minute per 100 liters.

The Sr-90 concentration was an average of 10 disintegrations per minute per 100 liters in the zone from 5°N to 25°S.

Migration of Ce-144, Y-91, Nb-95 and Tc-99 depends largely on the radionuclide content. It is almost impossible to guess which forms correspond to their physicochemical equilibrium in sea water. Specific laws of adsorption, colloid formation and other processes begin to affect the behavior of trace amounts of the substance in solution. The sea medium itself, which contains dissolved and colloidal organic matter, bacteria, marine hydrobionts and suspensions of different composition and origin, plays an important role.

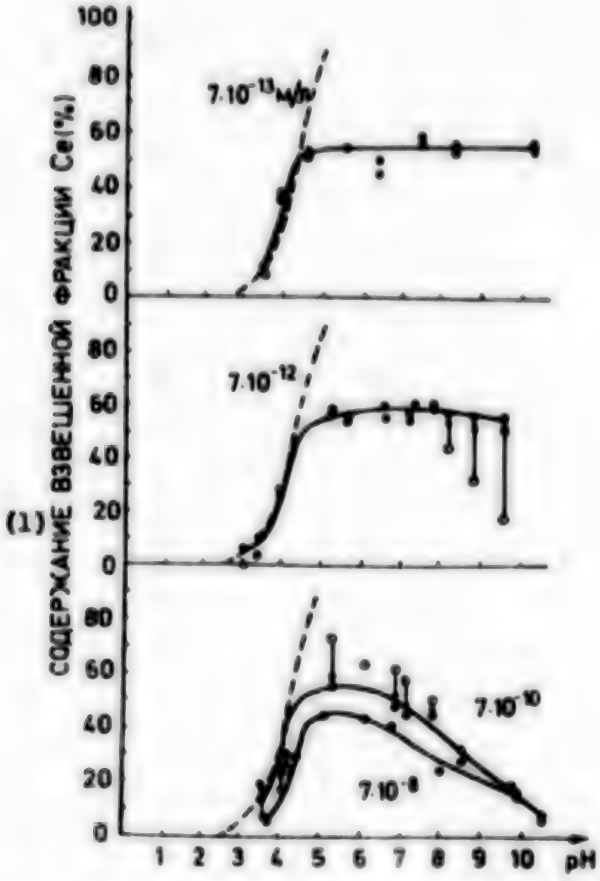

Figure 6. Effect of pH of Solution and of Total Concentration of Element on Formation of Suspended Ce-144 Fraction in Bidistilled Water

Key:

1. Content of suspended Ce fraction (percent)

Laboratory experiments were conducted in "pure" form on bidistilled water. The actual behavior was investigated on samples of sea water. Radioactive elements were introduced to the liquid phase in ion-soluble form.

Ultrafiltration and centrifuging of the solutions and also ion exchange and electrophoresis were used to study the state of radionuclides in the liquid phase. The fraction of the radioactive element that passed through the filter during ultrafiltration or that remained in the upper part of the beaker during centrifuging was regarded as dissolved (provisionally). The remaining part of the element was related to the "suspended" fraction. Ultrasmall quantitites of the elements in the solutions could in turn be found in the ion dispersion, molecular, truly colloidal and pseudo-colloidal states [21, 22].

Investigation of the Ce-144 distribution among dissolved and suspended fractions in bidistilled water as a function of the pH of the solution and concentration in the range of $7 \cdot 10^{-13}$ M to $7 \cdot 10^{-10}$ M showed that cerium remains almost entirely in solution in acid solutions at pH 5 at the given microconcentrations (Figure 6). According to [3], trivalent cerium cannot form truly colloidal $Ce(OH)_3$ particles which could be held on an ultrafilter under these conditions at pH = 9.3. Tetravalent cerium, which yields a colloidal fraction of hydroxide at pH = 3.9, also apparently does not form under the given conditions [24]. If $Ce(OH)_4$ were present in the investigated system, colloid formation at high values of pH would be significantly more intensive. The dashed lines of Figure 6 correspond to the theoretically calculated process of trivalent cerium hydroxide formation [25]. As can be seen from Figure 6, calculations and experimental data agree only on the initial segments of the curves which reflect formation of a suspended fraction. This indicates the occurrence of pseudo-colloids in formation of which dissolved silicic acid apparently plays a role [26]. A decrease of the amount of suspended fraction with a 100-fold increase of cerium ion concentration is apparently determined by processes of ion polymerization. Y-91, Nb-95 and Tc-99 also do not go completely into suspension in bidistilled waters at any values of pH [27-29]

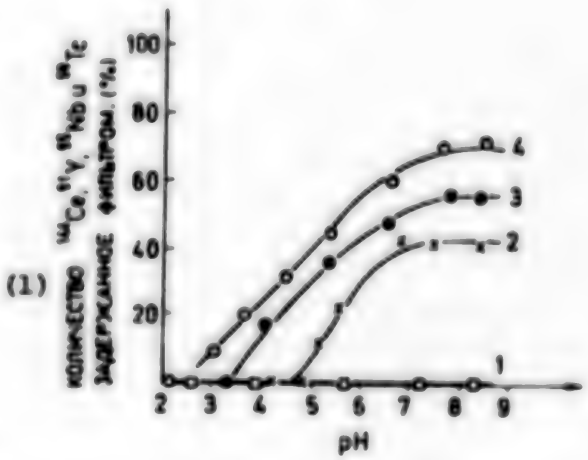

Figure 7. Variation of Quantity of Element Held by Ultrafilter as a Function of pH of Sea Water: 1--Tc-99; 2--Y-91; 3--Ce-144; 4--Nb-95

Key:

1. Amount of Ce-144, Y-91, Nb-95 and Tc-59 held by filter (percent)

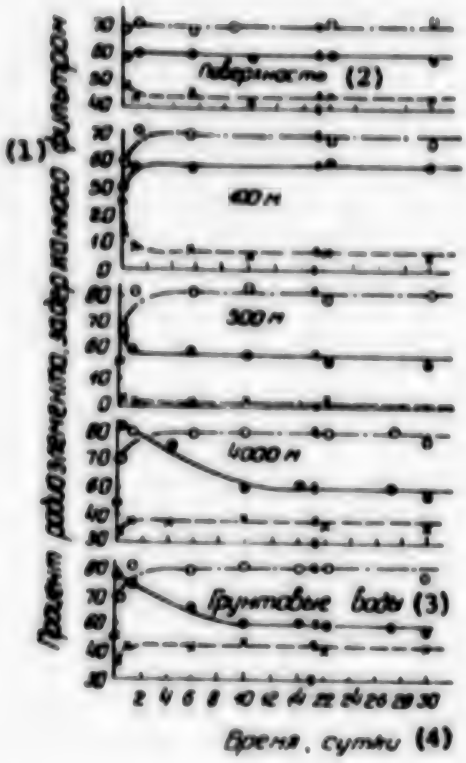

Figure 8. Formation of Suspended Ce-144 Fraction (Solid Lines), Y-91 Fraction (Dashed Lines) and Nb-95 Fraction (Dot-Dash Lines) With Time in Waters Sampled at Different Levels

Key:

1. Percentage of radioactive element held by filter
2. Surface
3. Bottom water
4. Time, days

Besides the hydrolytic properties of the element, its state is affected by adsorption processes in suspensions and complexing with organic ligands in sea water. The composition of the suspension and organic matter is not identical at different points and at different depths in the ocean. Therefore, an element may have different migration capability at different stages of its presence in the ocean. To ascertain this, the transformation of ionic forms of Ce-144, Y-91, Nb-95 and Tc-99 introduced to water samples taken from idfferent levels in the Pacific Ocean at the location of 00°56'N and 160°29'W was studied [30-31].

The hypothesis according to which rare-earth elements (RZE), falling to the sediment on the ocean bottom, can be returned to the water mass due to the effect of complexing with organic phosphorus-containing compounds, was also checked [32]. The behavior of Ce-144, Y-91, Nb-95 and Tc-99 in muddy water obtained from the surface layer of bottom deposits at a depth of 5,173 meters at the location indicated above was considered for this purpose.

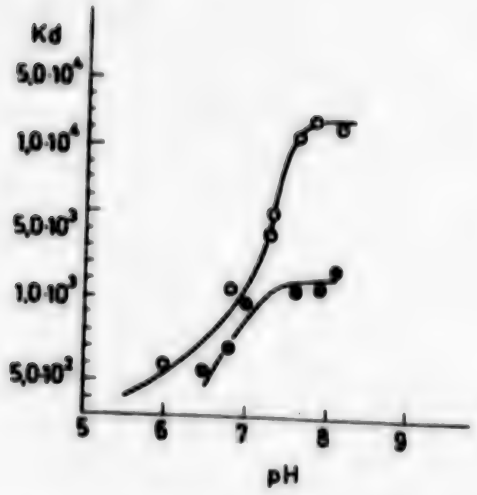

Figure 9. Equilibrium Values of Ce-144 and Y-91 Distribution Coefficients Upon Absorption of Them on Dauex-1 Anion Exchanger from Sea Water: 1--Ce-144; 2--Y-91

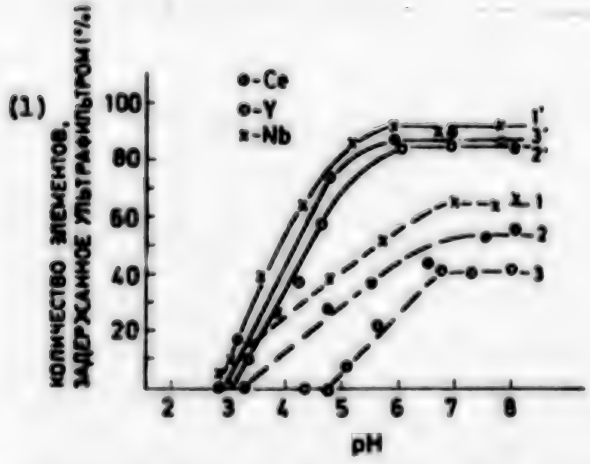

Figure 10. Effect of Iron on the State of Radioactive Elements in Sea Water: dashed lines--natural sea water; solid lines--sea water with addition of 10 micrograms per liter

Key:

1. Amount of elements held by ultrafilter (percent)

Some results of experiments obtained by the ultrafiltration method are presented inFigure 7. One may note that formation of the suspended fraction develops less intensively with an increase of pH than in bidistilled water. Technetium remains in the initial ionic state (ammonium pertechnate)

in the entire range of acidity of the medium. As in pure water, Ce-144, Y-91 and Nb-95 do not go completely into suspension at any values of pH. It was also shown that a reduction of the fraction of suspended Ce-144 is not observed at pH $>$ 7 and concentration greater than $7 \cdot 10^{-12}$ in sea water. This is apparently related to inhibition of polymerization of hydrolyzed ions by foreign impurities in sea water. Another reason is also possible. According to [33], oxidation of Ce^{+3} to a state of +4 is possible in sea water at pH $>$ 7. Because of this, a true colloid of difficult to dissolve cerium compounds, for example, the hydroxide or phosphate, should be formed.

The dependence on the acidity of sea water for Y-91 and Nb-95 is similar to that observed for Ce-144.

The results of investigating the distribution of the studied radioactive elements in water sampled from different depths are presented in Figure 8. The Ce-144 and Nb-95 content in the suspended fraction is essentially identical for water from all depths. The suspended Y-91 content is minimum (0-10 percent) in water from depths of 100 and 500 meters, while it is significantly higher (40-50 percent) in water from the surface, from a depth of 4,000 meters and in water from bottom deposits. Y-91 apparently interacts with some complex-forming agents whose concentration increases from the surface to a specific depth and then decreases again. These are possibly the products of decomposition of organic matter. The difference in the fate of Ce-144 and Nb-95, compared to Y-91, should result in differentiation of these nuclides upon transfer of them to the ocean depths and to faster settling of the former.

Ce-144 and Y-91 distribution between the suspended and dissolved fraction was essentially identical upon separation by ultrafiltration and centrifuging. The latter method permitted calculation of the mean radius of colloidal particles, which is in the range of 0.44-0.05 micron for Ce-144 and 0.20-0.05 micron for Y-91.

The results obtained by the ion exchange method for solutions in bidistilled water easily supplement each other and correspond to theoretical concepts of the interaction of pseudo-colloidal particles with anion and cation exchange resins [23, 34]. The absorption of the investigated radionuclides on a cation exchanger is essentially equal to zero at high values of pH.

Cerium and yttrium from sea water are absorbed only on an ion exchanger (Figure 9). Eighty percent of the Ce-144 and 30 percent of the Y-91 goes from solution to the anion exchange resin with a value of pH = 8.0, typical for natural sea water. Partial absorption of these elements on the anion exchanger and the absence of it in the cation exchanger are apparently related to formation of negatively charged particles during hydrolysis. The difference in separation of the investigated elements by ultrafiltration (by centrifuging) and by means of ion exchangers is apparently related to

formation of colloidal aggregates with sea water phosphates and soluble complex compounds with organic ligands, besides negatively charged particles. The results are in good agreement with data of electrophoretic investigations which indicate the appearance of negatively charge particles at pH > 7.5, which have a positive charge in the range of pH < 7.

Thus, upon entering sea water, the investigated radioactive elements are hydrolyzed with formation of negatively charged colloidal particles (pH = = 7.8-8.2). Moreover, Y-91 apparently forms neutral charged particles with phosphates and soluble complex compounds with organic ligands.

One of the constituent parts of the organic suspension is living matter which continually removes chemical elements from sea water and brings them into the biological circulation system. Another important component of the ocean's suspension is iron and its content in suspended matter fluctuates from 0.5 to 19.5 percent [35], 36]. Yielding a hydroxide, it may participate in extraction of elements from sea water.

Therefore, the effect of iron on the state of Ce-144, Y-91, Nb-95 and Tc-99 in sea water was investigated (by the ultrafiltration method).

The results are presented in Figure 10, from which it is obvious that Y-91, Ce-144 and Nb-95 go into suspension by almost 90 percent upon introduction of an additive quantity of iron. There is a total of 45 percent Y-91, 60 percent Ce-144 and 70 percent Nb-95 in the suspended state in natural sea water. The presence of iron does not effect the physicochemical state of Tc-99. The data clearly indicate the danger which results in disposal of radioactive elements in the ocean. Entering plankton organisms during feeding, iron serves as its own kind of carrier of fission fragments and contributes to implication of the investigated radioisotopes in the active biological cycle.

It was established that the coefficient of accumulation of all the isotopes in the presence of iron by plankton is an order higher than in the sea water-plankton-isotope system. The effect of the specific composition of the plankton was not loaded in this case.

Besides biogenic processes affecting the distribution of radioactive elements in sea water, abiogenic processes--uptake of isotopes by the suspension surface and bottom sediments--were also investigated.

With regard to the fact that the composition of the suspended matter is not uniform in depth, it was of interest to study the uptake of radioactive isotopes by suspension from different depths of the ocean. These samples were taken at depths of 0, 100, 500 and 4,000 meters at the location indicated above and also from the surface in different areas of the tropcial part of the Pacific Ocean. It was found that, regardless of the geographic location of the sample, the amount of Ce-144, Y-91, Nb-95 and Tc-99 extracted by suspension from the water is identical almost everywhere (Table 5).

Table 5. Uptake of Investigated Radioactive Elements by Surface Suspension

№ образца взвеси (1)	Середина района сбора пробы (2)		Захват радиоэлемента, K_d (5)			
	широта (3)	долгота (4)	^{91}Y	^{95}Nb	^{99}Tc	^{144}Ce
1	22°42'N	170°34'З	$4{,}1 \cdot 10^3$	$5{,}8 \cdot 10^3$	0	$1{,}9 \cdot 10^3$
2	12°29'N	156°36'З	$4{,}3 \cdot 10^3$	$5{,}6 \cdot 10^3$	0	$1{,}9 \cdot 10^3$
3	21°00'С	173°04'З	$4{,}5 \cdot 10^3$	$6{,}1 \cdot 10^3$	0	$1{,}9 \cdot 10^3$
ст. 5892 (6)	0°50'С	160°29'З	$4{,}3 \cdot 10^3$	$6{,}2 \cdot 10^3$	0	$1{,}7 \cdot 10^3$

Key:

1. Number of sample of suspension
2. Middle of sampling region
3. Latitude
4. Longitude
5. Uptake of radioactive element, K_d
6. Station

Study of the uptake of radioactive elements sampled at different depths by suspension showed that this process is quantitatively dependent on the properties of the suspension and its granulometric composition. It was noted for the investigated radioactive elements that the minimum uptake is confined to the surface suspension and it increases in proportion to the sampling depth. The maximum value was determined for the deep-sea suspension where mineral grains are a significant part of its composition. The irreversibility of radioisotopes taken up is also noted at these levels.

Phenomena of repeated dissolution and mineralization of suspensions and biological processes participate actively in migration of radioactive elements taken up by suspension. Being taken up by newer and newer plankton organisms as a result of frequent replacement of their generations, the radioactive elements remain in the biological sphere and are transferred from lower organisms to higher organisms, creating immediate danger to man, despite the opinion of the limited dilution of radioactive elements in the ocean.

More widely distributed and typical oozes sampled in the Indian and Pacific Oceans were taken to study the effect of bottom sediments on the distribution of radioactive elements in the ocean. The results of quantitative extraction of the investigated radioisotopes from sea water by bottom sediments showed that Ce-144, Y-91 and Nb-95 take up more completely silt which is contained in the thin fractions of more than 50 percent kaolite. A somewhat lower percentage of isotopes (approximately 85) is related to tropical Radiolaria by finely aleuritic silt containing more than 16 percent amorphous silica in the form of skeletal remains of plankton organisms --Radiolaria and partially diatom algae. Lesser extraction of isotopes is typical for slightly calcareous clay oozes (80 percent). Sediments with high calcium content (approximately 60 percent) and volcanic sand (approximately 75 percent) take up radioisotopes worst of all.

Thus, the investigated bottom sediments can be arranged in the following order by the extent of uptake of isotopes from sea water: noncarbonaceous clay ooze > tropical finely aleuritic Radiolarian ooze > slightly calcareous clay ooze > volcanic sand > carbonaceous ooze.

The data clearly indicate that the degree of uptake of elements by bottom sediments is a direct function of the surface area and the chemical composition of the latter. For example, one of the good concentrators of isotopes--finely aleuritic ooze--has very developed surface of 32.5 m^2/g and contains 16.3 percent amorphous silica and 1 percent calcium carbonate, while carbonaceous ooze, which is related to worse absorbers, has a surface of only 0.61 m^2/g, there is no amorphous silica in its composition and it consists almost entirely of calcium carbonate (93.5 percent).

Thus, as a whole the extraction of radioactive elements from sea water by bottom sediments is a complex physicochemical process which is a combination of the process of mechanical uptake of colloidal particles of these elements by oozes and of absorption, the part of the radioisotope which is in sea water in a truly diluted state.

The results outlined here in combination with data of [5-7] provide a pattern of radioactive contamination of the oceans in 1966-1967. The initial situation for further study of the radioactivity of one or another section of the ocean is given for the first time by each investigated region.

By 1966-1967, radioactive contamination of the surface waters of the ocean was reduced as a whole and some previously existing radioactive areas had become diffused. The Sr-90 content, which decreased with depth, clearly exceeded the amount in the Pacific Ocean which was deposited due to global radioactive fallout. Similar reliable data were found only at some points in the Atlantic Ocean.

The main circumstance which causes alarm is the presence of technogenic radioactive regions in international waters and the tendency toward an increase of their number, the intensity of contamination, dimensions and also toward fusion of these regions with each other.

To understand what may occur with the sea and its resources in the future, the physicochemical and biological processes which control radionuclides discharged into the ocean are being studied. Hydrolysis, complexing and concentration in the ocean suspension play an important role for the soluble forms of some radioactive elements. The implication of artificial radionuclides in the active biological circulation may result in accumulation of them in some links of the migration chain. Among the different types of bottom sediments, the best absorber of the investigated radionuclides is noncarbonaceous clay ooze. The mechanism of extracting these nuclides from sea water is a combination of mechanical uptake of colloidal particles of these materials by bottom sediments and by absorption on them

of the part of radioactive elements which is in sea water in a truly dissolved state.

It follows from the data given here that radioactive waste disposal in the oceans presents a specific danger. Taking into account the vigorous development of the nuclear industry and power engineering, effective measures must now be found to prevent further radioactive contamination of the oceans and seas.

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BIOLOGICAL EFFECTS AND BEHAVIOR OF RADIOACTIVE FISSION PRODUCTS IN AGRICULTURAL CHAINS

New York City PROCEEDINGS OF THE FOURTH INTERNATIONAL CONFERENCE ON THE PEACEFUL USES OF ATOMIC ENERGY in Russian Vol 11, 1972 pp 663-674

[Article by Ye. A. Fedorov, B. S. Prister, G. N. Romanov, N. I. Burov, B. A. Ukhanova, G. G. Ryabov and Ye. R. Ryabova, USSR State Committee on the Use of Atomic Energy, Moscow]

Abstract

[Text] This paper examines the agricultural aspects of the radiation situation which may arise when the environment is contaminated with a mixture of products of nuclear fission of different ages and with neutron activation of the soil. Experimental data obtained by conducting experiments under natural conditions on experimental plots are presented. The paper establishes a connection between the density of radioactive contamination of agricultural lands with a fresh mixture of nuclear fission products, the amounts of radionuclide accumulation in harvests of agricultural crops contaminated during different stages of development, the doses of gamma- and contact beta-irradiation and the radiation injury to young crops caused by these doses. The authors determined the value of the correlation coefficients of intensive one-time and continuous fall-out of radioactive fission products and their concentration in crops as a function of the composition of the radionuclide mixture, the stage of development of the plants at the time of the fall-out and the period elapsed since the contamination of the plants.

In field experiments the authors established the values of the correlation coefficients of the density of soil contamination by radioactive fission products, products of neutron activation and fissible materials and the amounts of their accumulation as a function of soil properties and of the biological characteristics of the plants. The role of the soil and foliar paths of intake by plants in contamination of crops by radioactive fission products and neutron activation is discussed.

In experiments involving the administration of a fresh mixture of products of ^{235}U nuclear fission to lactating cows, the authors studied the mechanisms of accumulation in the organism and elimination of radionuclides of iodine, tellurium, molybdenum, barium and others and calculated correlation coefficients for intake of the mixture by the animals and the radionuclide content of products of livestock-raising. A relationship was established between the intake of the mixture of nuclear fission products by cows in feed, the dose load on the critical organs and the degree of radiation injury to the animals. On the basis of the results presented in the report, the authors calculated the minimum density of radioactive contamination of plantings of agricultural crops, meadows and pastures with a mixture of radioactive fission products of different ages which cause radiation injury to pastured animals and plants.

Introduction

A program of wide development of atomic energy and use of atomic explosions for peaceful purposes is planned by many countries of the world. During the operation of enterprises of the atomic industry and atomic energy installations and also in the conducting of underground atomic explosions, some quantities of radionuclides may enter the environment. Overestimation of the danger of radioactive contamination of the environment, however, may lead to unjustified increases in expenditures for the construction and operation of installations and equipment for recovery of radioactive effluent. In this connection it becomes necessary to substantiate scientifically the levels of entry of radionuclides into the environment at which there is no radiation danger for man, animals or plants. The solution of this problem requires investigation of the quantitative relationship between the levels of the entry of radionuclides into the environment and their content in agricultural products.

The high reliability of atomic energy installations still does not eliminate the likelihood of accidents (1-5). In order to evaluate the consequences of radiation accidents and develop a system of special measures, it is necessary to establish the critical levels of the radionuclide content of the environment at which injury to agricultural plants and animals is possible.

During the past two decades extensive information has been accumulated concerning the behavior of radionuclides in the human environment (4, 6, 7). The results of investigation of this problem have been published in the materials of previous Geneva conferences and in the reports of the UN Scientific Committee on the Effects of Atomic Radiation (8, 9, 10).

At the same time many aspects of this problem require further study. This particularly applies to evaluation of the possible contribution of different paths of entry of radionuclides into agricultural products and also to prediction of their concentration in these products as a function of the level of the entry of radioactive substances into the environment.

The questions of the biological effect of the products of nuclear fission on plants and animals have not been sufficiently studied.

The present report discusses the results of observations of global fall-out and also of prolonged field and laboratory experiments studying the migration of radionuclides in biological chains in radical and foliar paths of entry into plants and the biological effect of fission products on farm animals and agricultural plants. In conducting the experiments we used solutions of nitrates of individual radionuclides and mixtures of new products of ^{235}U fission 10-hours old, which were administered to animals in their feed or sprayed on plots 50-200 m^2 in area using special devices.

The Entry of Radionuclides Into Agricultural Products During Radioactive Fall-out

Foliar entry of radionuclides into agricultural plants

A considerable amount of the radionuclides which fall out on the surface of the Earth fall directly on plants, bypassing the soil. In this case the radionuclide content in a crop is many times greater than with entry of the radionuclides from the soil through the root systems at one and the same fall-out density. In the case of continuous fall-out the radionuclide content of plants is determined by the rates of fall-out during the vegetative period; therefore it is advisable to attribute the radionuclide content of products not to total fall-out per year but to the average monthly intensity occurring during the vegetative season (Table 1). From the data presented in Table 1 it can be seen that with global fall-out of long-lived products the greatest radionuclide content is characteristic of agricultural products, which surpass milk and meat more than ten-fold in terms of ^{90}Sr and ^{137}Cs concentrations. Of the radionuclides examined, ^{137}Cs was distinguished by the greatest mobility in the food chain.

Table 2 presents data on the radionuclide content of wheat and potato crops in one-time contamination of plantings at different stages of development by a mixture of ^{235}U fission products 10 hours old. As our investigations have shown, the major factors which determine the intensiveness and amounts of incorporation of radionuclides from fall-out in plants are the species characteristics of plants, the developmental stage at the moment of contamination and the chemical nature of the radionuclides. The influence of the indicated factors on the amounts of contamination is so significant that at an equal density of radionuclide fall-out at different times in the vegetative period their content in agricultural products may differ many hundred-fold.

The most significant factors are the species differences and the stage of development of the plant during the period of fall-out. The maximum content of radionuclides in the crop is observed when the plantings are

contaminated during the period of formation of the productive organs when radionuclides intensively move into them with the flow of nutrient substances. The intensity of the movement of radionuclides into plants is to a significant degree determined by the presence of stable nuclides--chemical analogues which play a specific role in the mineral nutrition of plants.

When plantings of field crops are contaminated with a mixture of new fission products, a considerable amount of short-lived radionuclides disintegrate by the time the crop is consumed, and basically only the long-lived radionuclides ^{90}Sr, ^{137}Cs and certain others may be present in the crop. The productive organs of vegetables are consumed in fresh form in food without any significant extraction or treatment, with the result that along with milk, vegetables may be the basic supplier of the short-lived and biologically extremely dangerous radionuclides of iodine, molybdenum, barium and others.

Table 3 presents data concerning the content of these nuclides in harvests of widely distributed species of vegetable crops. The productive part of most vegetables is the vegetative organs, and for this reason the radionuclide content of vegetables is significantly greater than in the productive part of harvests of field crops.

From the data of Table 3 it can be seen that the radionuclide content of the vegetable harvest after 10 days was 2-7-fold lower than at the time of application of the radioactive solutions. Such a significant decrease in the radionuclide content is basically explained by losses of them by the plants and to a lesser degree by dilution through increase in the biomass. Long-term investigations established that the process of loss of radionuclides by plants is described by an exponential relationship. For most nuclides and plants studied the length of the period of half-loss is 0.5-3.0 days when the solutions were administered during the stage of intensive biomass development and from 7 to 100 days during the period of ripening (11).

Assimilation of radionuclides from the soil by agricultural plants

Regardless of the duration of fall-out, in the final analysis radionuclides fall on the soil surface, penetrate the root layer and become available for assimilation by the root systems of plants. Table 4 presents the amounts of root assimilation of radionuclides (fissible materials, the products of their fission and the products of neutron activation of soils and structural materials). Radionuclides can be arranged in the following series according to capacity for accumulation from the soil by plants:

Content in Plant pCi/kg product / uCi/m^2	$n \cdot 10^4$	$n \cdot 10^3$	$n \cdot 10^2$	1-10	$n \cdot 10^{-1}$-10
Radionuclides	^{65}Zn	^{185}W	^{137}Cs, ^{90}Sr, ^{59}Fe	^{144}Ce, ^{106}Ru, ^{95}Zr	^{239}Pu, ^{235}U, ^{147}Pm, ^{91}Y

In root assimilation from the soil the amounts of accumulation of radionuclides in agricultural crops is basically determined by the chemical condition of the nuclides in the soil and by the nature of their reaction with the soil. The role of stable nuclide-analogs in physiological processes is also of important significance to the processes of radioisotope assimilation.

Correlation of the data presented concerning the radionuclide content of harvests of agricultural crops (Tables 1-4) demonstrates what an important role the foliar path of entry of radionuclides into plants plays. Thus, the radionuclide content of wheat grain is tens of times higher with the foliar path of entry than in the case of assimilation from the soil at an equal ultimate density of soil contamination. This difference is many times greater for the vegetative organs of the plants. At the same time the radionuclide content of potato tubercles which have not been subjected to direct contamination by fall-out is practically identical with both the radical and foliar paths of entry.

The transfer of radionuclides from feed to the products of livestock-raising

Plants are the basic source of entry of radionuclides into animals and the products of livestock-raising. Table 5 presents the maximum concentrations of radionuclides in the milk, muscles and liver of cows after one-time entry of them into the organism in a mixture of fresh fission products. The results presented correlate well with the data of other investigations (4, 12).

Knowing the values of the maximum concentration of radionuclides in milk and of the effective periods of their half-life it is possible to predict the nuclide content of milk and meat products at different times after one-time consumption of contaminated feed. In terms of their concentration in milk and meat the radionuclides under investigation form a descending series $^{131}I > ^{99}Mo > ^{89}Sr > ^{140}Ba$. Abnormally high accumulation is observed in the liver, apparently caused by the participation of Mo in the oxidizing-regenerative processes of the organism.

The above examined experimental data on the behavior of radionuclides in food chains (soil--plants and fall-out--plants--animals--products of

livestock-raising) may be used in order to determine the safe levels of entry of radionuclides into the environment. On the basis of data on the consumption of basic food products by the population of the USSR and the limits for annual intake (PGP of radionuclides through the digestive organs established by the Norms for Radiation Safety (NRB-69) in force in the USSR, we calculated the maximum levels of radionuclide fall-out and content in the soil of agricultural lands (Table 6). Analysis of the published data (1, 5) shows that the levels of radionuclide fall-out in regions where operating atomic energy stations are located are significantly lower than the values presented in Table 6.

The Effect of Ionizing Radiation on Agricultural Plants and Farm Animals

The effect of radiation on agricultural plants

A number of investigations have shown (1, 3) that the radiation doses to the plants and animals in the vicinity of atomic energy installations is considerably lower than the critical values. In accidents, however, the density of radioactive fall-out may prove to be so great that it causes radiation injury to biological subjects.

Up to the present a certain amount of information has been accumulated on the effect of chronic and acute exposure on the growth and development of vegetating plants (4, 13, 14) obtained in experiments on gamma-fields. For purposes of predicting the loss possible with fall-out of radioactive fission products on plantings of agricultural crops it is necessary to have data on the relation between the size of the irradiation dose of plants and animals and the density of the contamination of the territory.

Field experiments with artificial contamination of plantings of wheat, barley and potatoes with a mixture of 10-hour-old products of ^{235}U and ^{90}Y fission, the physical characteristics of which were close to those of a mixture of fresh fission products, made it possible to establish the quantitative relationship between the density of radioactive contamination of plantings and the size of the dose absorbed by the plants.

It was established that the degree of radiation injury to cereals is basically caused by the size of the dose absorbed by the most radiosensitive organ--the growth cone or the forming ear. The ratio of the doses of beta- and gamma-radiation absorbed by these organs is 30-80. Owing to the small penetration capacity of beta-particles the size of the dose of beta-radiation absorbed by the critical organs of the plant depends on the stage of its development during the exposure period.

It is possible to distinguish three stages in the growth and development of plants; these are distinguished by the size of the dose absorbed by the critical organ. The first period--from the beginning of germination to the end of tillering--is characterized by the fact that the growth cone is located under the soil surface, is not exposed on the daytime surface and is essentially shielded from beta-radiation. The second

period--from the beginning of tillering to spike formation--is characterized by the underground location of the critical organ, which is shielded from beta-radiation only by the stem tube and the leaves adjoining the stem. The third period--from the putting out of the ear to complete ripening of the grain--is distinguished from the previous periods by the fact that in this case the critical organ is the germ of the forming grain, which is shielded from beta-radiation by the flower and ear scales.

In one-time contamination of plantings with a mixture of 10-hour-old nuclear fission products with a density of 1 Ci/m^2, the size of the dose absorbed by the critical organ is 0.1, 10 and 60 krad for periods 1, 2 and 3 of development, respectively.

Suppression of growth and substantial (as much as 50 percent) decrease in the grain yield of wheat and barley are observed when the plants are irradiated during periods 1 and 2 with doses of 1.5-3.0 krad. Complete sterility occurs when the size of the absorbed dose is 12-15 krad. The results obtained confirm the results of experiments involving irradiation of vegetating plants with gamma-sources (13, 14).

The third period is characterized by the least radiosensitivity. Thus, in the experiment with barley when the size of the dose absorbed by the ripening grain was greater than 60 krad the yield during the year of irradiation decreased by no more than 10-15 percent.

Analysis of the results presented shows that plantings of cereals are most sensitive to contamination during the period from the beginning of tillering to spike formation. The size of the dose absorbed by the critical organ of the plant which causes injury to the plantings is 3.0 krad for this period; this corresponds to a fall-out density of 300 mCi/m^2 for the mixture of new fission products. The size of the critical absorbed dose for other agricultural crops is determined by their relative radiosensitivity. According to the results of our investigations and the data (13) the critical dose for legumes is not significantly different from the doses for cereals; injury to plantings of potatoes, for which the period from the beginning of tubercle formation is critical, can be expected at a density of contamination with a mixture of new fission products of 600 mCi/m^2.

The Effect of Radiation on Animals

The radioactive contamination of plantings and pastures in radiation accidents may result in the entry of considerable amounts of fission products into animals. Of interest in this connection is study of the behavior of radioisotopes in animals and the possibilities of injury to dairy cattle, since it is known that milk and milk products are an important component of the human diet, and cows may reduce or lose milk productivity long before the dose of external irradiation reaches a lethal size.

A number of investigators have studied the biological effect of external gamma-irradiation on dairy cattle (12, 15). With intake of a mixture of fresh fission products by cows, however, the dose distribution is distinguished by great heterogenity in comparison with external irradiation. This circumstance makes it impossible to apply the results of experiments on external irradiation of animals directly to the case of internal irradiation occurring with the intake of fission products with the feed.

A study of the distribution of the absorbed doses in lactating cows which have ingested fresh fission products was conducted in tests on 10 animals. A mixture of 10-hour-old fission products was administered in the feed a single time and once a day for four days.

Table 7 presents the values of the absorbed doses in the critical organs of the experimental animals. The greater part of the absorbed dose (80-90 percent) is formed during the first three days and therefore the nature of the irradiation corresponds to acute irradiation. The gastro-intestinal tract and thyroid gland are subjected to the greatest exposure; the disturbance of their normal functions as the result of accumulated radioactive iodine results in reduced lactation. This makes it possible to infer that reduction in milk productivity is one of the most sensitive indices of radiation injury in ingestion of a mixture of fission products.

The disturbance of the function of the thyroid gland which accompanies reduced milk yield may occur with exposure to a dose of more than 3 krad (16). According to the data of Table 6 exposure of the thyroid gland to such a dose is possible when approximately 100 mCi of a mixture of fresh fission products is ingested. It can be inferred that ingestion of a mixture of fresh fission products by cows in a quantity exceeding 100 mCi may result in a reduction in milk productivity.

The data examined make it possible to evaluate the levels of radioactive contamination of pastures in one-time fall-out of a mixture of new fission products which may result in loss of milk productivity. On the average the daily ration of cows is 50 kg of fresh grass, which corresponds to the consumption of vegetation from an area of 160 m^2 (4). The value of the coefficient of initial retention of radionuclides may be assumed to be equal to 25 percent (11). Then an overall intake of 100 mCi of fission products will occur when the density of the contamination of the pasture is equal to approximately 2 mCi/m^2.

The data examined concerning radiation injury to agricultural plants and farm animals when lands are contaminated with nuclear fission products show that injury to pastured animals may occur at a considerably lesser density of radioactive fall-out than injury to crops.

Table 1. The Radionuclide Content of Agricultural Products in Global Fall-Out*

$$\frac{\text{nCi/kg of air-dried matter}}{\text{mCi/km}^2\cdot\text{month}}$$

(1) (2) (3) (4) (5)

Радионуклиды	Сено трав	Пшеница(зерно)	Молоко	Мясо
^{90}Sr	4,1	0,34	0,03	0,08
^{106}Ru	2,0	0,14	-	-
^{137}Cs	5,0	0,52	0,17	0,54
^{144}Ce	3,3	0,03	-	-

*The average values in three years, $\sigma \leq 50\%$.

Key:
1. Radionuclides
2. Hay
3. Wheat (grain)
4. Milk
5. Meat

Table 2. The Radionuclide Content of Harvests of Field Crops After a Single Contamination of Plantings With a Mixture of 10-Hour-Old ^{235}U Fission Products*

$$\frac{nCi/kg\ product}{\mu Ci/m^2}$$

Радионуклиды (1)	(2) Фаза развития в момент загрязнения					
	(3) Пшеница (зерно)				(4) Картофель (клубни)	
	3-4 листа (5)	Выход в трубку (6)	Молочная спелость (7)	Восковая спелость (8)	Активное цветение (9)	Увядание ботвы (10)
$^{89,90}Sr$	0,19	1,1	57	2,1	1,0	0,02
^{137}Cs	0,35	1,0	37	1,1	0,17	0,06
^{95}Zr	0,31	2,6	18	1,5	6,7	0,09
^{140}Ba	0,02	0,44	2,8	2,1	0,07	0,006
$^{141,144}Ce$	0,30	0,22	0,2	6,6	0,41	0,11
$^{103,106}Ru$	0,09	0,19	1,5	7,1	0,23	0,12

*The average values in three years, ± 30%.

Key:

1. Radionuclides
2. Stage of development at the time of contamination
3. Wheat (grain)
4. Potato (tubercle)
5. 3-4 leaves
6. Stem formation
7. White ripeness
8. Yellow ripeness
9. Active flowering
10. Withering of plant tops

Table 3. The Radionuclide Content in Harvests of Vegetable Crops 1 and 10 Days After a Single Contamination

nCi/kg product
$\mu Ci/m^2$

Культура (1)	^{99}Mo		^{131}I		^{140}Ba	
	(2) 1 день	(3) 10 дней	1 день	10 дней	1 день	10 дней
Лук	240	40	280	110	130	17
Капуста, листья	440	52	770	130	430	62
Редис	480	96	530	160	370	100
Салат	490	140	800	310	450	84
Шпинат	600	300	940	320	500	100

$\sigma \approx \pm 30\%$

Key:

1. Crop
2. 1 day
3. 10 days
4. Onions
5. Cabbage, leaves
6. Radishes
7. Lettuce
8. Spinach

Table 4. Radionuclide Content in Harvest After Administration of Them in Soluble Form[a, b]

$$\frac{\text{pCi/kg product}}{\mu Ci/m^2}$$

(1) Радионуклид	(2) Пшеница (зерно)	(3) Картофель (клубни)	(4) Сеяные злаковые травы (сено)
^{90}Sr	300	270	1200
^{137}Cs	66	370	74
^{144}Ce	1,9	11	3,5
^{106}Ru	7,4	10	7,6
^{147}Pm	0,09	1,0	0,9
^{90}Y	0,1	24	0,33-19
^{95}Zr	11	13	18-270
^{65}Zn	$3,6 \cdot 10^4$	$1,7 \cdot 10^4$	$0,9 \cdot 10^4$
^{185}W	460	1100	1200
$^{55,59}Fe$	250	42	58
^{239}Pu	0,02-1,5	2,5	1,0
$^{238,235}U$	1,4	1,3	2,9

a Average value in three years,
b Soil is leached, fertile chernozem; the content of metabolic Ca is 44 milliequivalents

Key:
1. Radionuclide
2. Wheat (grain)
3. Potato (tubercles)
4. Sown cereal grasses (hay)

Table 5. Maximum Concentration (C, $\frac{nCi/kg}{\mu Ci}$ of Radionuclides in Milk, Meat and Liver of Cows in A Single Ingestion in Feed and the Effective Periods of Half-Life of the Nuclides (T_{eff}, day)

(1) Продукты	(2) Молоко		(3) Мясо		(4) Печень	
(5) Радионуклиды	C	(6) $T_{эф}$	C	(6) $T_{эф}$	C	(6) $T_{эф}$
^{131}I	13	1,0	0,40	1,5	4,0	0,41
^{99}Mo	0,9	0,78	0,32	0,59	93	0,23
^{89}Sr	0,3	2,0	0,012	2,2	0,004	2,9
^{140}Ba	0,05	0,92	0,003	2,0	0,016	4,2
^{132}Te	-	-	0,022	0,57	4,3	0,32

σ ≤ ± 20%

Key:
1. Products
2. Milk
3. Meat
4. Liver
5. Radionuclides
6. T_{eff}

Table 6. Maximum Levels of Radioisotope Fall-Out and Soil Content Agricultural Lands At Which the Annual Human Intake of Radionuclides Does Not Exceed the PGP

Радионуклид (1)	Содержание радионуклидов в почве[a], Ки/км² (2)	Интенсивность непрерывных выпадений[b], мКи/месяц·км² (3)
^{90}Sr	2	1,5
^{137}Cs	250	50
^{131}I	-	0,5[c]

a Chernozem soils; the critical products are bread and milk.
b Average zone of the USSR; critical products are bread and vegetables.
c $\mu Ci/km^2 \cdot day$

Key:
1. Radionuclide
2. Radionuclide content of soil[a], Ci/km^2
3. Intensiveness of continuous fall-out[b], $\mu Ci/month \cdot km^2$

Table 7. Absorbed Doses in Critical Organs of Cows After Ingestion of a Mixture of 10-Hour-Old Products of ^{235}U Fission in Feed (mrad)

	Поглощенная доза (1)	При однократном поступлении 1 мКи (2)	При 4-кратном (раз в сутки) поступлении, когда величина первой порции равна 1 мКи (3)
(4)	Средняя тканевая γ-излучения	15	35
(5)	β-излучения в тонком отделе кишечника	50	70
(6)	β-излучения в толстом отделе кишечника	100	140
(7)	β-излучения в щитовидной железе	$12 \cdot 10^3$	$35 \cdot 10^3$

Key:

1. Absorbed doses
2. After single ingestion of 1 mCi
3. After fourfold (once daily) ingestion, when the size of the first portion equals 1 mCi
4. Average tissue gamma-radiation
5. Beta-radiation in small intestine
6. Beta-radiation in large intestine
7. Beta-radiation in thyroid gland

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MIGRATION OF RADIONUCLIDES IN FORESTS, AND EFFECTS OF IONIZING RADIATION ON TREE PLANTATIONS

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[Text] Introduction

The study of the patterns of distribution and migration of artificial radioactive substances in various elements of the environment and of the effects of ionizing radiations on communities of plants and animals is a task that is important in both the practical and theoretical respects. Tree biogeocenoses occupy a special place among natural communities. Forests as a landform, that occupies a significant part of land, have a strong influence on the distribution and migration of radioactive substances in the biosphere on a global scale, preventing development of processes of radionuclide transport by wind and water over the earth's surface, while the cycle of synthetic radionuclides in them is characterized by unique distinctions that are specific only for forest cenoses.

The forest is a system that is very sensitive to ionizing radiation; it is damaged by radiation in doses that do not induce noticeable disturbances in other types of phytocenoses, in particular in communities of herbaceous plants. For this reason, the study of the patterns of migration of artificial radionuclides in forests and effects of ionizing radiation on tree cenoses is of practical interest, from the standpoint of both evaluation of radiation effects when radioactive substances are discharged into the environment and investigation of the possibility of commercial utilization of tree products with high radionuclide content. In addition, the study of processes of migration of radionuclides in forests opens up the possibility of gaining fuller knowledge about the mechanisms and patterns of biogeochemical processes and the cycle of chemical elements in tree biogeocenoses, while the study of radiation effects will definitely aid in gaining deeper understanding of the structural and functional distinctions of forest cenoses as self-regulating systems.

In this paper, we submit the results of experimental studies of forest radioecology, which were pursued for many years in the USSR and directed toward two main objectives: analysis of the patterns of distribution and migration of radionuclides in forest cenoses, and study of the consequences of irradiating forests, which are comprised of effects attributable directly to radiation and secondary phenomena related to changes in structure and biogeocenotic relations in an irradiated cenosis [1]. These results sum up the information obtained from a series of experiments involving introduction of radionuclides in forest plantations by means of spraying the crowns with radioactive solutions and putting them under the canopy, as well as the data obtained from observing distribution and migration of synthetic radionuclides in forests after global and one-time local fallout.

Migration of Radionuclides in Forests

Primary interaction between tree plantations and radioactive fallout: When transported by the wind, the radioactive particles are trapped the most effectively by trees standing on the edge of a forest. According to our observations, 2-5 times more radionuclides settle on the crowns of forest edge trees than the fallout on adjacent treeless areas; the "edge [of forest] effect" is demonstrable at a distance of 15-20 m from the edge of the forest.

For quantitative description of interaction between radioactive fallout and vegetation the coefficient of retention is generally used; it refers to the share of radionuclides retained by the above-ground part of plants. The magnitude of this coefficient is related to the type and age of the tree stand, seasonal and meteorological conditions, as well as physicochemical forms of nuclides. According to our observations, the coefficient of retention fluctuates over the following range, depending on specific conditions:

Pine seedlings, 6-10 years old, crown density 1.0	Crowns sprayed with ^{89}Sr solutions	90-100%
Pine plantation, 60 years old, crown density 0.9	Fallout of particles up to 50 μm in size	80-100 %
Pine plantation, 25 years old, crown density 0.8	Fallout of particles up to 100 μm in size	70-90%
Pine plantation, 30 years old, crown density 0.8	Fallout of secondary (soil) particles raised from the surface by wind	40-60%
Birch plantation, 40 years old, before leaves open, crown density 0.8	Fallout of secondary (soil) particles raised from the surface by wind	20-25%

Thus, tree vegetation is characterized by a higher retention capacity with respect to radioactive fallout than herbaceous vegetation (for which the coefficient of retention constitutes a mean of 25% [2]. This is attributable to the large biomass of the crowns, better separation thereof and high ratio of leaf and needle surface to weight thereof, for which reason the canopy plays the role of a filter that is capable of retaining a significant amount of radioactive dust. Since dry radioactive fallout hits the ground in relatively small amounts by weight, which are not sufficient to saturate the crowns completely, the coefficient of retention of radioactive fallout by the forest canopy can apparently be considered to equal the degree of compactness [density] of crowns, with the exception of forests of deciduous trees, during the period they are without leaves. In this case, the forest canopy has a retention coefficient that is about one-third the quoted figure.

Cycle of radionuclides in forests: Vertical and horizontal migration begin immediately after radionuclide fallout, under the influence of the wind and atmospheric precipitation, as well as shedding of trees, as a result of which the radionuclides gradually migrate from the top parts of the crowns to the lower ones and then under the canopy.

Seasonal conditions, the role of which we demonstrated in experiments involving spraying pine seedling crowns with ^{89}Sr solutions, are an important factor that affects the rate of removal of radionuclides from crowns. When ^{89}Sr was applied to the crowns in the summer (June), they were relatively rapidly cleared of it, in spite of the dry weather (3 mm rain in the first 2 weeks). The half-life for removal of ^{89}Sr as related to the entire above-ground biomass, constituted about 3 weeks under such conditions. When ^{89}Sr was applied in the fall (October), the rate of self-purification of crowns was significantly lower: half-life for elimination from the crowns as a whole (without counting radioactive fallout) constituted about 4 months, i.e., 5.5 times longer than when used in the summer (Figure 1).

The experimental data pertaining ot distribution of radionuclides penetrating into tree crowns in the form of solid particles indicate that removal of contaminants from the above-ground part of the tree stand also depends, in this case, on the time of year when the fallout occurred. Thus, according to our observations, when there was radioactive particle fallout in the spring, the half-life for migration from the crowns of pine tree stands constituted 4 months, versus 10-12 months in the case of autumn fallout (Figure 2). Thus, elimination of fallout from the above-ground part of adult tree stands is 10-12 times slower than for herbaceous vegetation, for which the half-life for elimination of the most important fission products contained in radioactive fallout constitutes 12-14 days [3].

The rate of migration of radionuclides under the canopy of crows in the case of fall and winter fallout depends on the type of forest. According to our data, in deciduous forests, over 50% of the radionuclides entering the crowns migrate to the forest floor after the leaves fall in autumn.

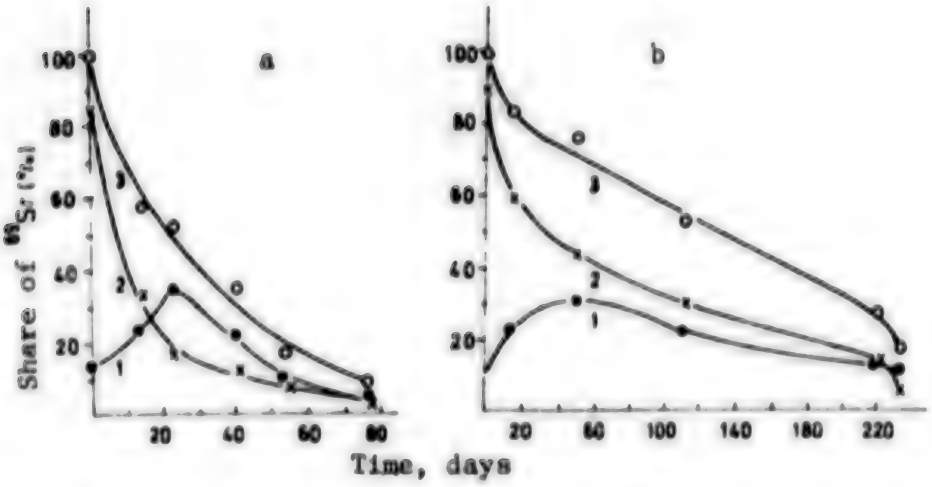

Figure 1. Change in ^{89}Sr content of pine crowns when the radionuclide is used in the summer (a) and autumn (b) (% of total levels) in: 1) branches, 2) needles and 3) needles + branches

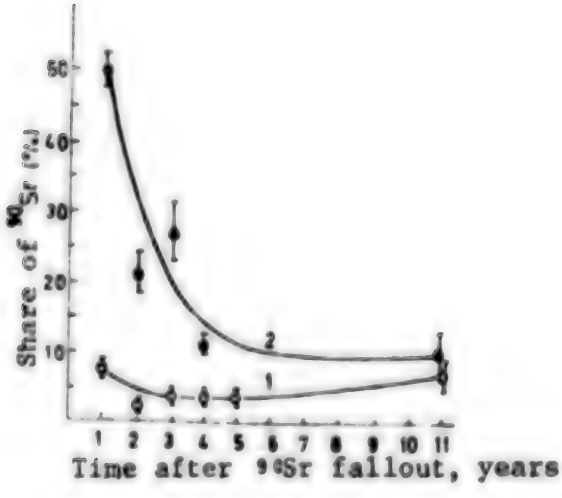

Figure 2.
Changes in ^{90}Sr content in aboveground part of birch (1) and pine (2) forests (% of total level in cenosis) after single fallout

Concurrently with vertical migration of radionuclides from the crowns into the ground, there is redistribution over the level [height] of the crowns. The radionuclides pass from the overstory to the lower part of of the crown canopy, where the effect of rain and wind is less marked, as a result of which the radionuclide content in the bottom part of the crowns is several times higher than in the top part. After a certain time, most of the radionuclides are concentrated in the forest floor and soil. This initial stage of redistribution of radionuclides in the forest cenosis is largely irreversible. Experimental studies of the cycle of ^{90}Sr over a period of many years in adult birch and pine forests after single fallout of this radionuclide revealed that this stage, during which up to 90% of the radionuclide migrates under the forest canopy, lasts about 1 year in birch plantations and 3 to 5 years in conifer forests, and this is related to the longer life of pines and rougher surface of the branches (Figure 3).

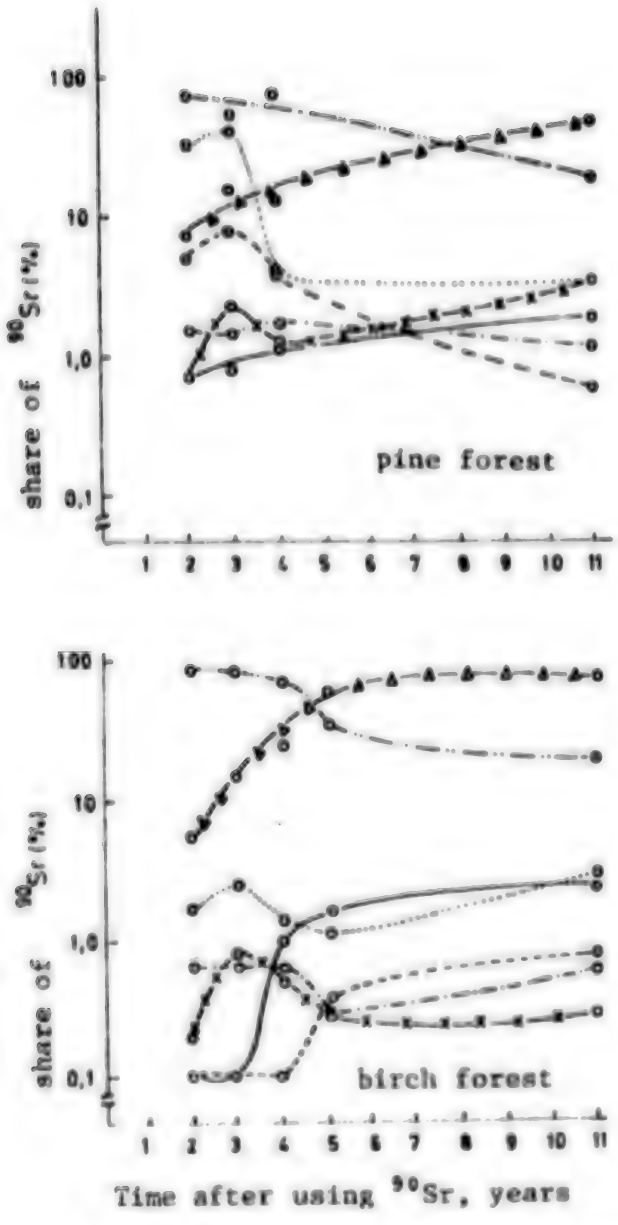

Figure 3.
Changes in ^{90}Sr content in different components of forest cenosis (% of total) after single fallout
0) experimental data
..) forest floor
) soil
-) wood pulp
-.-) bark
---) leaves (needles)
...) branches
x) grassy vegetation

After this stage, the soil becomes the main source of radionuclides in the above-ground part of trees. According to our observations, the period during which 50% of ^{90}Sr fallout in the course of migration of radionuclides under the forest canopy and mineralization of the forest floor migrates into the soil and becomes accessible to assimilation by the roots constitutes about 4-5 years for the birch forest and 8-9 years for the pine forest, which is attributable to differences in rate of self-purification of crowns and rate of mineralization of the forest floor in deciduous and coniferous forests. To forecast the maximum levels of accumulation of radionuclides in the above-ground perennial parts of trees, we considered the fact that upon reaching equilibrated distribution, the ratio of radionuclide concentration in the biomass to its concentration in soil should equal the corresponding ratio of concentrations of stable isotopes of a given element:

$$\frac{C_{rad.\ plant}}{C_{rad.\ soil}} = \frac{C_{stab.\ plant}}{C_{stab.\ soil}}$$

A good conformity was obtained of estimated concentrations with the actual maximum concentrations of ^{90}Sr in wood pulp, which are reached in the pine and birch forests 5 years after radioactive fallout (Figure 4) [4].

The distribution of radionuclides in a forest cenosis is close to a state of relative dynamic equilibrium 10 years after radioactive fallout, in which the annual transfer of radionuclide from the soil to the above-ground part exceeds the reverse

migration only by the amount corresponding to the increment in the current year of this nuclide in the biomass.

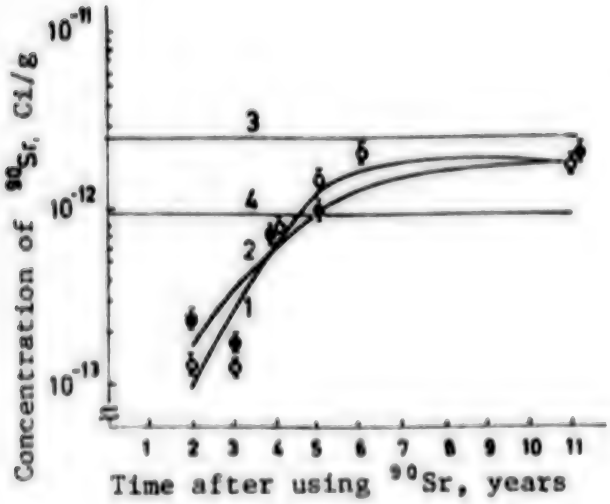

Figure 4.
Accumulation of ^{90}Sr in wood pulp of birch trees (1) on chernozem and pine trees (2) on podzol soil. ^{90}Sr content in cenosis 1 mCi/m^2. 3, 4--estimated maximum concentrations of ^{90}Sr in birch and pine pulp, respectively

Dosimetry of Ionizing Radiation in Forests After Radioactive Fallout

When there is radioactive fallout in forests, the highest doses of radiation are received by the assimilating organs of trees and tissues of the apical meristem. On the one hand, this is due to the fact that these organs are open and most of the radionuclides settle in them. On the other hand, the size of these organs is commensurate with the range of β-particles emitted by fission products, and all of the cells of these tissues are accessible to γ-radiation; most of its energy is absorbed in them. For this reason, although β-radiation does not play an appreciable role in external irradiation of the cambial meristem and dormant buds, which are protected from radioactive fallout by a layer of bark, it makes the main contribution to the radiation dose to leaves, needles and apical meristem tissues.

In order to determine the correlation between radionuclide content of crowns and radiation doses, special experiments were conducted, with spraying of ^{89}Sr solutions on the crowns [5]. The doses of β-radiation were measured with photodosimeters placed on different parts of the crowns. The doses absorbed within the meristematic tissues (particularly the "soil" [probably typo for buds]) were estimated on the basis of distribution of the radionuclide in such tissue, the dosage to the bud surface being normed to the dose measured by the photodosimeters. Thus, when scaled to 1 mCi ^{89}Sr/m^2, the total dose constituted about 170 rad in needles and 90 rad in bud meristem when the radionuclide was sprayed in the summer, 550 and 300 rad, respectively, i.e., 3.2 times more, when used in the fall.

The reduction of absorbed dose in bud meristems is due to the fact that these tissues are significantly protected against external β-radiation by the bud scales; only a negligible part of the radionuclide applied penetrates into the buds through the scale surface.

Radiation Damage to Trees by Radioactive Fallout

The main patterns of effects of ionizing radiation on trees in cases of radioactive fallout have much in common with the effect on tree plantations of very penetrating radiation (γ-quanta, neutrons) from point sources, which has been described comprehensively on the basis of research conducted in the United States involving exposure of some segments of a forest to radiation [6, 7]. At the same time, there are some specific distinctions to radiation damage to trees from radioactive fallout, which are related to the distribution of radiation sources, which changes in time and space, and consequent changes in dose fields in a forest biogenocenosis.

Effect of radioactive fallout on woody plants: Radiologically, woody plants are the most vulnerable component of a forest cenosis with regard to radioactive fallout. On the one hand, this is due to the great intensity of β-radiation in the crowns at the early stage and, on the other hand, the relatively high radiosensitivity of trees, particularly conifers. We studied the radiation effects of β- and γ-radiation of radionuclides falling on tree crowns the most comprehensively in common pine (Pinus silvestris) and common birch (Betula verrucosa). As in the case of external γ-radiation, they present a decrease in growth increment, inhibition of needle and runner growth, dying off of needles and buds and formation of new buds and runners, double increment, impairment of monopodial branching, phenological changes manifested by delayed opening of leaves in stricken trees.

The specifics of radiation damage to trees caused by fallout refer to the fact that when radionuclides fall in the form of relatively large particles (up to 100 μm), the damage to leaves, needles and tissues of the apical meristem is manifested primarily in the lower and upper parts of the crowns, particularly on the windward side. The top runners remain viable when up to 95% of the crown dies, and the apical meristem of the main runner remains intact, even though it is among the most radiosensitive tissues of the tree. This is attributable to relatively rapid decontamination of the apical runners under the influence of the wind and atmospheric precipitation. Unlike this, with exposure of trees to external γ-radiation from point sources, the apical meristem is the first to be injured, and the crown starts to dry at the apical runner.

A comparison of resistance of conifers and deciduous trees to fallout revealed that the differences in resistance to summer fallout are mainly attributable to species-specific differences in radiosensitivity (birch

trees are 5-7 times more resistant than pines). But if fallout levels are taken as a criterion of resistance, the differences between pine and birch are even greater (by about 20 times) in the case of fallout in the autumn and winter, because of the decreased retention of fallout (and consequently radiation dose) in the crowns of deciduous trees after they have shed their leaves.

Resistance of woody plants to radiation depends significantly on the physiological state. Thus, when ^{89}Sr is sprayed on pine crowns when the trees are in a state of physiological dormancy, lethal doses of radiation (absorbed dose in needles 3200 rad) were 1.6 times higher than when the radionuclide was applied during the vegetation period (2000 rad). According to Sparrow [8], the seasonal differences in radiosensitivity of trees are attributable to an increase in volume of meristem cell nuclei (by about 1.6 times) when they make the transition from winter dormancy to active growth.

The ratios we obtained between radionuclide content of the crown and β-radiation dose rate, and our comparison of the observed radiation effects to dosage enabled us to determine the overall radiation damage to forests after fallout from a mixture of fission products. According to our data, the contribution of β-radiation to the total dose accreted in leaves and needles is several times greater than that of γ-radiation, since most of the energy of β-radiation of radionuclides in the crowns is absorbed in the crown biomass, whereas the share of energy from γ-radiation absorbed by the above-ground part of the tree stand constitutes no more than 10% of its total energy. The total energy of β-radiation from a mixture of fission fragments at the age of up to 100 days is close to the total energy of γ-radiation, and for an older mixture it could be several times greater.

Thus, our results are indicative of the important role of β-radiation in radiation damage to forests. For example, in the case of fallout on a forest from a fresh mixture of fission products during the vegetation period, the ratio of contributions of β- and γ-radiation to the dose absrobed in leaves, needles and apical meristem tissues may reach 9:1, and with increase in age of the mixture the contribution of β-radiation would be even greater. If, however, the fallout occurs in late autumn or winter, the fallout levels that kill woody plants will be considerably higher (by 1.5-2 times for evergreen conifers). The seasonal differences in lethal levels of fallout would apparently be even greater (5-6-fold) for woody plants that shed their leaves (or needles) in the winter, since most of the radionuclides from winter fallout will directly go under the forest canopy without being retained in the crowns.

Effect of radioactive fallout on organisms under the forest apron: After radionuclides migrate under the apron, they concentrate in the thin layer of forest floor, creating rather high doses of radiation in it and adjacent layers of soil and air. This is precisely the area where plant seeds fall

and where numerous species of various animals live--worms, insects, Arachnida and small mammals (myomorphs and insectivores). Experiments dealing with the effects of ^{90}Sr and ^{90}Y β-radiation on conifer seedlings--pine, fir and larch--revealed [9] that irreversible damage to seedlings of this species begins with an absorbed dose rate of 10-15 rad/day, which corresponds to 6-9 mCi $^{90}Sr + ^{90}Y/m^2$ in the top layer of soil. With such levels, seedlings cease to grow and perish in the third year of their life. With an absorbed dose rate of 4 rad/day (3 mCi $^{90}Sr + ^{90}Y/m^2$) radiation, no irreversible damage is caused, although there is a decline of growth rate. In the fourth year of life, the seedlings begin to show satisfactory growth.

When the fallout levels are high enough and the crowns of trees are damaged by radiation, the direct effect of ionizing radiation on organisms living under the forest canopy is combined with the effect of altered microclimate (increased light, elevation of temperature of top layer of soil, etc.) due to thinning of the crowns under the effect of radiation. For this reason, the resultant effect of radioactive fallout on the population of these organisms is determined not only by their radiosensitivity, but adaptability to altered environmental conditions.

Let us consider the changes in composition of grass cover after fallout of long-lived radionuclides on a forest as an illustration of the combined effect of these factors. There was a 3-5-fold increase in grass cover biomass in plantations where pine trees died of irradiation and birch trees were severely damaged (30% of the trees dried up). At the same time, there was a change in species composition of the association. According to the data of Ye. G. Smirnov, bush grass (Calamagrostis epigeios), a plant capable of vegetative reproduction from rhizomes protected by a layer of soil from β-radiation, became the dominant species in the association 2-3 years after the fallout. This species was also rather widespread under the canopy of the control plantation. As time passed, the following plants also became dominant: Chamaenerium angustifolium, Centaurea scabiosa and Cirsium setasum, which are photophilic tall species, and pioneers with a powerful root system that eject radioresistant but relatively short plants, such as Fragaria vesca, Carex precos and a few others.

The long-term presence of populations of plants and animals in an environment with a high background of ionizing radiation triggers adaptation mechanisms directed toward adjustment of organisms to these altered conditions. Increase in radioresistence (radioadaptation) is one of the manifestations of adaptation. According to our data, the incidence of chromosomal aberrations in seedling cells and young plants was lower after additional "stimulating [provocating]" γ-irradiation of seeds of plants that had been growing for 7-10 years in an area with high concentration of ^{90}Sr (0.3 mCi/m^2) than when seeds from plants in a control plot were irradiated (Figure 5). It can be assumed that, in the case of chronic exposure to a high background of ionizing radiation, there is elimination

from a population of organisms that is heterogeneous in radiosensitivity of the more radiosensitive specimens and selection of the more radioresistant forms.

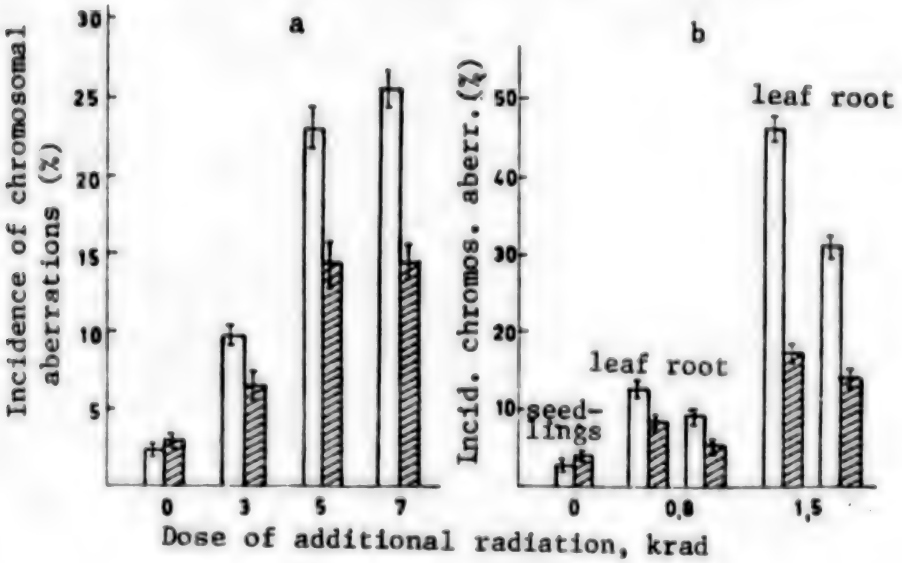

Figure 5. Change in incidence of chromosomal aberrations as a function of dosage of provocating radiation in plants from control (white columns) and experimental (striped) plots

a) Vicia crocca L. (seedlings) b) Agrimonia eupatoria L.

Direct irradiation and alteration of the structure of the forest cenosis and microclimate as a result of radioactive fallout have an appreciable influence on forest fauna as well. These changes can be illustrated by the results of our observations of the gipsy moth (Lymantria dispar), which is a widespread primary pest in forests of the temperate zone (Figure 6).

Exposure of eggs to chronic radiation in doses of $n \cdot 10^1$-$n \cdot 10^2$ rad stimulated hatching of caterpillars, while higher doses ($n \cdot 10^2$-$n \cdot 10^3$) caused appreciable decrease in hatching. Radiation did not have an adverse effect on survival of caterpillars living in the same parts of the forest, since the radiation dose to the crowns, where the caterpillars feed at this stage, were significantly lower than near the surface of the ground. Moreover, we found that the higher the radionuclide content in a given area, the higher the survival rate of caterpillars. This is attributable to the decrease in number of tachina flies, which parasitize on gipsy moths, under the influence of radiation. The high sensitivity of tachina flies to radioactive fallout is attributable to the fact that they hibernate on the

forest floor (at the pupa stage), where rather high doses of radiation are generated (up to 30 rad/day absorbed dose rate) and where they undergo metamorphosis.

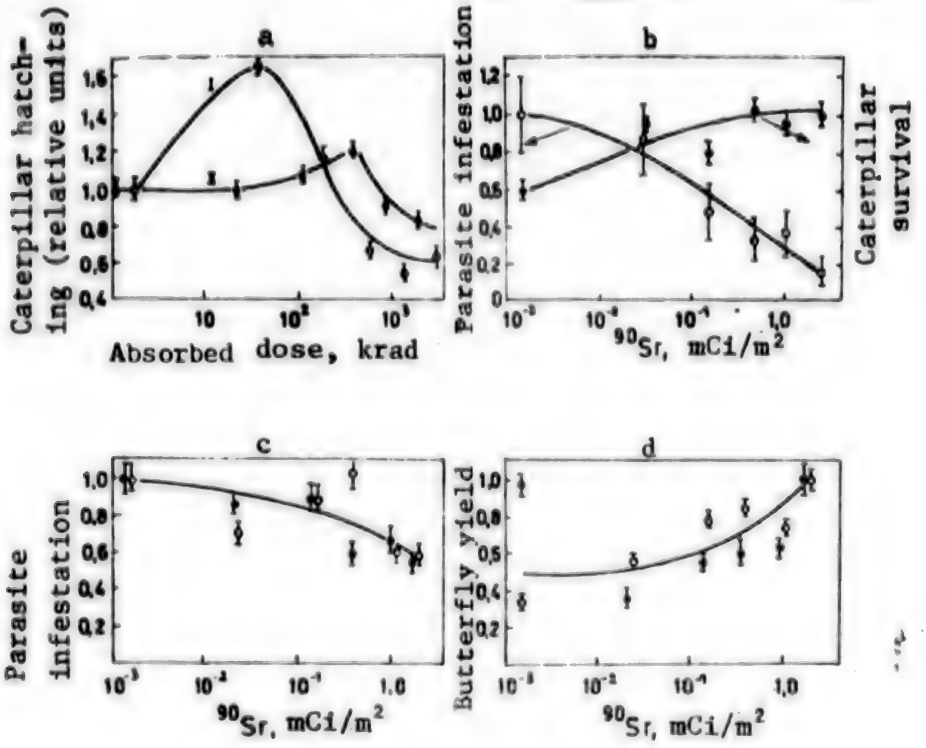

Figure 6. Effect of single ^{90}Sr fallout on population of gipsy moths and their parasites, tachina flies

a) caterpillar hatching as a function of radiation dose
b) infestation of caterpillars by tachina fly larvae and caterpillar survival as a function of level of ^{90}Sr fallout
c) infestation of pupae by tachina fly larvae
d) yield of butterflies as a function of level of ^{90}Sr fallout
ϕ ϕ) experimental data for two successive generations of gipsy moths

There is an analogous, though less marked, correlation betw tent of parasite infestation and survival of the "host" as a functic fallout level in the gipsy moth at the pupa stage. In addition to irradiation and parasite activity, other factors (change in influx of heat and light under the forest canopy, increased airing of crowns, impairment of food chain manifested by impaired coordination of leaf opening and caterpillar hatching, etc.) have an appreciable influence on size of gipsy moth population. We used the number of eggs laid per unit area as an integral criterion reflecting the combined effect of all factors influencing the number of gipsy moths in forests damaged by radiation. We found that there was a decrease in eggs laid with increase in fallout level, and

this decrease is unrelated to a decrease in amount of food, since there was less damage by caterpillars to leaves in areas with high radionuclide content than those with a low one, although the store of food per oviposition was significantly lower in the former case.

Changes in populations of herbivorous myomorphs [mouse-like rodents] could be another example of the effect of radioactive fallout on animals in forests. As a result of development of a good grass cover under the canopy of the damaged plantation, favorable conditions are created for mass scale reproduction of the rodents, which can survive in the presence of a rather high radionuclide content in the forest. Thus, Il'yenko [10] established that introduction of ^{90}Sr under the forest canopy, in doses of 0.6-3.4 mCi/m^2, did not have an appreciable influence on the life span of small rodents. Radiation only led to an increase in variability of morphological, craniological and other features of the populations, which expands the potential adaptability of the populations to existence in areas with a high levels of ionizing radiation.

Recovery of forests: The capacity of tree stands to recover from damage caused by ionizing radiation depends on the type of forest and extent of damage. Under our experimental conditions, the plantations of deciduous trees--birch and aspen [sic?]--retained the capacity for recovery by forming root or stump shoots when the crowns of the trees had dried completely. Thus, in the area where 70% of the new growth and 30% of the overstory of birch trees had died as a result of radioactive fallout, profuse growth of stump shoots appeared after 1.5 years, which were quite viable and developed for many subsequent years. In the case of partial damage to wood plants, they recovered after 8-10 years, even after 95% of the crown had dried. Since the apical runners usually remained undamaged in such a case, after restoration of the crowns the appearance of these trees did not differ from normal ones.

Thus, the results of our studies indicate that forests, as a component of the landscape, are among the most sensitive biological systems with regard to radioactive fallout. Ionizing radiation excludes from the forest biogeocenosis the most radiosensitive species and subsequently impairs its structure, due to changes in biogeocenotic relations between the different elements of the cenosis. Studies of cenotic reactions to radioactive fallout are still at the first stage. Deeper knowledge of the patterns of distribution of radionuclides in forests and of the effects of ionizing radiation on forest biogeocenoses is the objective of future studies.

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PROSPECTS FOR EXTENSIVE PEACEFUL UTILIZATION OF ATOMIC ENERGY WITHOUT RADIATION RISK TO THE POPULATION

New York City PROCEEDINGS OF THE FOURTH INTERNATIONAL CONFERENCE ON THE PEACEFUL USES OF ATOMIC ENERGY in Russian Vol 11, 1972 pp 703-709

[Article by Yu. A. Izrael' and E. N. Teverovskiy, USSR Main Administration of Hydrometeorological Service, Moscow, RSFSR]

[Text] Energy, released during the transformation of atomic nuclei, is used more and more widely by man. Atomic energy is the main new trend in the development of energy. [1].

At present the centers of atomic energy are atomic electric power plants (AEP) which use U^{235} fission by fuel neutrons. The projected development of nuclear energy is related to fast neutron reactors using U^{238}.

It is expected that the general use of energy by the year 2000 will reach 2.1×10^{10} tons of conventional fuel and the capacity of electric power plants will increase to 1.1×10^{7} M W [3,2]. The percentage of atomic electric power plant capacity out of the total electric power plant capacity will increase from 1.3 percent to approximately 50 percent [2] and will reach 3×10^{6}, and according to some findings, 5.5×10^{6} MW [2,3].

The efficiency coefficient (e.c.) of the atomic electric power plants is now 25-30 percent, in the future it may increase to 42-45 percent [2].

In addition, in the coming years we can expect the development of new uses of atomic energy in industry and construction.

It is exceedingly important that the development of nuclear energy not lead to harmful or even dangerous effects as a result of the possible release of certain quantities of radioactive products of nuclear reactions into the environment. This factor can be a real (and perhaps, a decisive) influence on the development of nuclear energy as a whole as well as on the development of some of its branches. More attention is being paid to the problems of the effect of nuclear energy on the environment [3,4].

No any one branch of industry has such protective measures against possible environmental pollution as nuclear energy [3]. In spite of this, more and more effective measures are being implemented, possible emission doses in the immediate vicinity of the plants are being evaluated, the harmful effects of atomic plant emissions are being compared to that of plants operating on conventional fuel (even natural radioactivity is taken into account when calculating the emissions of such establishments, etc.) [3]. In some cases (in certain areas) the dose, resulting from natural radioactivity at conventional fuel plants is comparable and can even exceed the dose from radioactive products emitted by AEP's [5].

However, in order to evaluate the prospects of developing nuclear energy, evaluations on a global scale--extending over countries and continents--are necessary.

The significance of general evaluations of radioactive aftereffects of nuclear energy development and the comparison of the harmful effects of using conventional fuel with the possible harmful effects of the industrial use of nuclear energy is therefore understandable.

The harmful effects of using conventional fuel are generally related to the emission of harmful substances which are formed during the combustion of the fuel. In the case of electric power plants and motors (those which use coal, fuel oil, and other petroleum products) these are basically sulfur dioxide and ashes, hydrocarbons, nitrous oxide, lead monoxide, etc.

In the last years, some 200-250 million tons of fuel ashes and various types of dust and approximately 60 million tons of sulfur dioxide have been emitted into the atmosphere world-wide. The amount of toxic substances in the air of several large cities significantly surpasses the maximum possible concentration (MPC) [6].

The emission of sulfur dioxide in the burning of coal presently in 160-2040 g per 1 ton of burned fuel. The amount of ashes (taking into account ash collectors) can exceed the specified amounts by several times [6]. According to the data in article [6] it is possible to write an approximation formula for the amount of sulfur gas a (ton per 24-hour period) emitted by an electric power plant into the atmosphere during the combustion of fuel oil:

$$a \approx 0.09\, \rho \alpha \qquad (1)$$

where

ρ = the capacity of the plant in MW
α = the amount of sulfur in the fuel oil, in percent

If we take as the average value = 2.5 percent, then the annual emission of sulfur dioxide by conventional fuel electric power plants, assuming equal emission per unit of power and burning liquid and hard fuels, is (at a total

capacity of 10^6 MW) approximately 80 million tons, and in the year 2000, it will make up (at a total capacity of 5.5×10^6 MW [2]) approximately 450 million tons of SO_2 and approximately 1.5 billion tons of ashes. Of course, this total can be somewhat decreased by the application of very expensive purification methods.

The possible harmful effects or danger in the use of nuclear energy is related to the formation of radioactive products as a result of fission or fusion as well as to possible induced radiation caused by the interaction of neutrons with the nuclei of the elements of the various surrounding construction.

At present we have accumulated much experience with the functioning of nuclear reactors (not less than 10^3 reactor-years of work) [3].

During the working of the atomic electric power plant the fission products gradually accumulate in the active zone of the reactor; the construction of present day plants effectively excludes the emission of such products into the environment in the course of normal reactor function [4] (the problem of radioactive waste is not considered in this work). During this process only an insignificant amount of gaseous isotopes Ar^{41}, C^{14}, H^3, Xe^{133} and traces of I^{131} can fall into the atmosphere. Therefore, according to [5], some atomic reactors in the U.S. release up to 650 Ki/24-hours of Ar^{41}. According to [7] the radiation dose even in the zone of the plant will not exceed 1 percent of the permissible.

It should be mentioned that in the process of dissolving spent fuel elements in radiochemical production, the emission of Kr^{85} (the only long-living inert gas) into the atmosphere is apparently unavoidable. In that case, we should assume a formation of 4.2×10^5 Ki of Kr^{85} at complete fission of 1 ton of U^{235} (approximately per 800 MW annual electric energy production per AEP).

In the event of an accident at an AEP, it is possible that iodine isotopes and several other relatively volatile isotopes can be released into the atmosphere. The biggest emission in the history of reactor construction took place during an accident at Windscale, was accompanied by the burning of the active zone and consisted of 2×10^4 Ki of I^{131}; 1.2×10^4 Ki of Te^{132}; 6×10^2 Ki of Cs 137; 80 Ki of Sr^{89} and 2 Ki of Sr ^{90} [8].

According to the evaluation given in [9], the frequency of accidents on a reactor site is less than 10^{-2} per 1 reactor year. The probability of large emissions of I^{131} is exceedingly small. In the work [10] it is considered most probable that a large accident (accompanied by the emission of several thousand Ki of I^{131}) can happen once in 1000 years of reactor use. The curve which describes the possibility of the emission of another amount of I^{131} depending on the length of reactor use is described by the law $t^{0.6}$. The degree of danger--in a series of accidents--in the form of cancer of the thyroid gland in regions of great population density (up to 5000 people per km^2) is greatest during the emission of 10^3Ki of I^{131} and totals 3 cases [11].

The evaluation of radioactive conditions and the determination of danger (risk) in the use of atomic reactors can be made by taking into account experimental data in each individual case. The results of the calculations influence the selection of the site, the safety measures and the protective measures taken. In addition, in each case, the recommendations and standards (international or national) for the protection of the population against radioactivity should be considered, as well as the rational evaluation of the relationship between use and risk.

However, in the quick development of nuclear energy it is important to have a clear perception of the possibility of nuclear energy development keeping in mind the possible radioactive effects in the whole world as well as a prognosis of the global radioactive condition concerning all probable emissions when the developmental programs are actually put into effect. It is necessary to determine absolute amounts of radioactive products which are found in nature, the doses obtained from these products and to compare the possible effects of the given pollution with the effect of pollution by non-radioactive harmful substances.

In calculating the global radioactive condition in the development of nuclear energy we weill assume the following:

--by the year 2000 about 5×10^3 reactors with an average capacity of 10^3 MW each will be in use;

--the distribution of the reactors on the land masses will be more or less regular;

--the number of significant accidents with emissions of approximately 10^3 Ki of I^{131} is approximately 5 (one accident per 10^3 reactor-years), or smaller accidents with an emission of 10^2 Ki of I^{131}--about 30 per year.

Thus, depending on the age of the reactor, up to 5×10^3 Ki of I^{131}, 30-140 Ki of Cs^{137}, 4-7 Ki of Sr^{89} and 0.1-1.0 Ki of Sr^{90} will be released into the atmosphere in a year [8].

The isotopes mentioned above will for several days settle on the land masses (calculating the rate of effective settling equal to 0.1-1 cm/sec) and will not have managed to spread out over the globe. In the calculations it can also be assumed that all of these isotopes will settle either on dry land or in the zone of the crippled reactors (approximately 10^7 km^2 not considering possible overlapping.)

The most dangerous isotope in an accident (for respiratory ingression and ingression with milk) is I^{131}; the other isotopes represent a danger mainly as a source of external radiation (Cs^{137}) or by ingression with food or water (Sr^{89}, Sr^{90}). According to our calculations and the data in [8] the pollutants of the earth's surface which approach the limits of radiation doses for the population (per year) are: for Cs^{137}-1.0-2.0 Ki per km^2; for Sr^{90}-0.06-0.6 Ki/km^2, and for I^{131}-50 mKi/km^2 (for children).

During the functioning of the reactor, H^3 and C^{14} may enter the atmosphere. Tritium is formed in the reactor as a result of fission and neutron capture in deuterium and lithium 6 (approximately in equal quantities) and enters the atmosphere in quantities up to 10-30 Ki/MW annually [12].

In the dissolving of spent fuel, approximately 3×10^9 Ki of Kr^{85} per year can enter the atmosphere (according to the projected capacity levels of the AEP by the year 2000). Longliving tritium and Kr^{85} will mix equally in the lower layer of the earth's atmosphere. The dose of tritium is significantly less than that from Kr^{85} and consists of on the average 10^{-3} mR/year for the whole population by the year 2000.

The data for the various isotopes (most dangerous) averaged for local areas ($10^7 km^2$) is given in Table 1 in fractions of doses or permissible ingression for the population [14]. The data for Kr^{85} and H^3 are averaged on the basis of the whole earth.

ТАБЛИЦА 1. МАКСИМАЛЬНЫЕ ОСРЕДНЕННЫЕ ЗНАЧЕНИЯ КОНЦЕНТРАЦИЙ В ВОЗДУХЕ (ИЛИ УРОВНЕЙ ЗАГРЯЗНЕНИЯ) РАЗЛИЧНЫХ РАДИОАКТИВНЫХ И ТОКСИЧНЫХ ВЕЩЕСТВ ПРИ РАБОТЕ ОБЫЧНЫХ И ЯДЕРНЫХ ПРЕДПРИЯТИЙ (в долях предельных количеств предела дозы, предельно допустимых поступлений для населения или предельно допустимых концентраций, - осредненных за год)

3) Изотопы и вещества	1) Атомные электростанции					2) Обычные электростанции	
	4) Обычная работа		5) Аварийная ситуация			SO_2	6) Летучая зола
	^3H	^{85}Kr	^{131}I	^{90}Sr	^{137}Cs		
7) На земной поверхности				10^{-6}	10^{-6}		
8) В воздухе	$5 \cdot 10^{-6}$	$3 \cdot 10^{-3}$	$7 \cdot 10^{-4}$			10	10
9) Максимальная величина для данного вида энергетики	$3 \cdot 10^{-3}$		~10^{-3}				10

Table 1. Maximal Average Values of Atmospheric Concentrations (or Pollution Levels) of Various Radioactive and Toxic Substances During the Operation of Conventional and Nuclear Plants (in fractions of limit quantities: dose limits, population ingression limits or the limits of permissible concentrations - averaged for a year)

Key:

1. Atomic power plants
2. Conventional power plants
3. Isotopes and substances
4. Normal operation
5. Emergency situation
6. Volatile ash
7. On the earth's surface
8. In the atmosphere
9. Maximal value for the given type of energy

In the calculations we assumed that the distribution (intermixing) of the relatively short-lived isotope takes place in the lower 200-meter layer of the atmosphere. For the long-lived, Kr^{85} and H^3, we used the 500 meter layer

of intermixing, in view of their longevity in the atmosphere. In calculating the atmospheric pollution by conventional toxic substances we assumed their homogenous distribution also in a 200-meter lower layer of the atmosphere. Taking for SO_2 a life equal to 4 days [15] and the amount of expected emissions by the year 2000 to be 4.5×10^8 tons/year, we obtain that the air over the dry land will have on the average a constant ten-fold increase over the average daily allowable limit of 0.05 mg/m^3. Analogous figures are obtained for volatile ash. Close figures (in relation to MPC) are obtained in calculating the concentrations of other toxic substances.

The data given in Table 1 show that in the operation of conventional plants, 10^3-10^4 times more air is needed to dilute the toxic substances to acceptable levels than is required in the operation of atomic electric power plants (under the conditions described above). In addition, this relationship does not depend on the accepted height of the mixing layer as distinct from the values of exceeding permissible concentrations.

Although the limit levels of pollution for radioactive and non-radioactive toxic substances are based on somewhat different factors, nevertheless, we should keep in mind that emissions of toxic substances (radioactive and non-radioactive) can lead to direct harmful effects in part of the population as well as to genetic defects [16].

Let us also note that the use of atomic energy does not require the use of oxygen which is necessary for the combustion of conventional fuel, and it does not lead to a constant increase of CO_2 in the atmosphere.

Therefore, the conclusion can be drawn that the development of atomic energy ensures sufficient cleanliness of the environment. Moreover the substitution of atomic energy for energy obtained on the basis of conventional fuel will lead to a significant decrease in the pollution of the environment by toxic substances and will result in its eventual return to a healthier state.

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[317-9233]

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